Hyperfine splitting in $^{209}\text{Bi}^{80+}$, $^{209}\text{Bi}^{82+}$ and beyond


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The specific difference between the ground state hyperfine splittings (hfs) in hydrogen-like and lithium-like ions of the same isotope was suggested about fifteen years ago as the ultimate tool to prove bound-state QED in the strong magnetic field generated by the heavy nucleus [1].

The isotope of bismuth, $^{209}\text{Bi}$, exhibits both ground-state hyperfine transitions very close to the visible spectrum and therefore they can be probed by laser spectroscopy. The $1s$ hfs in hydrogen-like bismuth ($^{208}\text{Bi}^{82+}$) was measured by direct laser spectroscopy at the experimental storage ring (ESR) in 1994 [2]. Seventeen years later and using an improved laser spectroscopic technique [3] at the same ring we found the $2s$ hfs in lithium-like bismuth ($^{209}\text{Bi}^{80+}$) [4]. Combined with a new measurement of the $1s$ hfs in the hydrogen-like bismuth we found good agreement with the theoretical prediction. Yet the accuracy of our result was limited at that time by the calibration of the electron-cooler voltage, determining the velocity of the ions in the ring [4].

We have repeated this experiment in 2014. This time we were able to monitor the electron-cooler voltage in situ using a voltage divider provided by PTB Braunschweig. First results on the $1s$ hfs in hydrogen-like bismuth using a coating beam have been already published in [5].

After two years of studying potential systematic errors we have now achieved relative accuracies at the $10^{-5}$ level for both hyperfine transitions [6]. These are the most accurate transition wavelengths measured in a heavy highly charged ion so far. This improvement has finally allowed us to improve the accuracy on the specific difference by an order of magnitude. A significant fact is that our new result shows now a $7\sigma$-difference to the latest theoretical prediction [7]. Such a large discrepancy was not expected by theory and therefore it has put the specific difference into question as a tool to test QED in strong fields.

It has, however, been pointed out that the specific difference is still sensitive to the nuclear magnetic moment of $\mu(^{209}\text{Bi})$ [7, 8]. A small variation from the tabulated value could bring theory and experiment into agreement. Therefore, there is also a need to remeasure this ground state nuclear property. A new measurement of $\mu(^{209}\text{Bi})$ via nuclear magnetic resonance is in preparation at TU Darmstadt and on a long-term perspective, a measurement of the hfs in both ion species is planned at the SpecTrap Penning trap and of the magnetic moment directly on hydrogen-like bismuth at the Penning trap ARTEMIS, which are both installed at HITRAP/GSI.

In order to confirm both, the reliability of the proposed nuclear structure independence in the specific difference as well as any assumption of a different nuclear magnetic moment value, we are now considering to measure the hfs in hydrogen-like and lithium-like ions of a second isotope. Two candidates are envisaged: $^{207}\text{Bi}$ and $^{208}\text{Bi}$. Their magnetic moments are ascribed to the single proton outside the $^{209}\text{Pb}$-core and the additional neutron holes below the $N = 126$ shell-closure. The specific difference in $^{207}\text{Bi}$ is expected to have a deviation that scales with the magnetic moment compared to that in $^{209}\text{Bi}$ because of the similar magnetic moment and spin. In the case of $^{209}\text{Bi}$, a new measurement of the magnetic moment relative to $\mu(^{209}\text{Bi})$ has recently been performed at COLLAPS/ISOLDE by collinear laser spectroscopy [9]. If the moment of $^{209}\text{Bi}$ is really different from the literature value, then we expect a similar disagreement for the specific difference in the case of $^{208}\text{Bi}$. In case that our disagreement with theory is an artefact due to an incomplete cancellation of the Bohr-Weisskopf effect in the specific difference, the deviation for $^{208}\text{Bi}$ should not scale with the magnetic moment since a considerably different Bohr-Weisskopf effect is expected for this isotope with a different spin and nuclear magnetism distribution. Bound-state strong-field QED can only be proven if this cancellation works as proposed in [1].

References


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Asymmetries of the electron cusp in heavy-ion atom collisions∗†

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The well-known experimental technique of zero-degree cusp electron spectroscopy has been extended towards heavy-ion atom collisions at near-relativistic collision velocities, at which new effects of asymmetries in the electron cusp arise.

In collisions of heavy highly-charged projectile ions with atomic targets, the energy distribution of the emitted electrons is a characteristic observable for the underlying elementary charge-transfer processes [1]. At the experimental storage ring ESR of the heavy-ion accelerator facility GSI, a dedicated magnetic electron spectrometer was installed downstream from the supersonic gas-jet target, which enables the measurement of high-energetic electrons emitted in ion-atom collisions, with electron velocities similar to the projectile velocity, emitted within a small cone in the forward direction (Figure 1). This technique provides the ability to extend the well known study of zero-degree cusp electrons towards heavy-ion atom collisions at near-relativistic projectile energies.

Figure 1: Magnetic electron spectrometer at the ESR.

Through the electron-loss-to-continuum (ELC) cusp, double-differential cross sections of projectile ionization can be studied even for the heaviest few-electron projectiles [2]. But also a new channel opens up, the radiative electron capture to continuum [3], which can be directly compared to its non-radiative counterpart [4]. Using the electron spectrometer in combination with detectors for emitted x rays and charge-exchanged projectiles, the study of the collision system U^{90+}(1s^22s^2) + N_2 @ 90 MeV/u revealed all three processes, each characterized by a unique shape of the electron cusp [5].

Furthermore, the process of electron loss to continuum was investigated for multi-electron projectiles in the collisions of U^{26+} with gaseous targets of H_2, N_2, and Xe at collision energies of 30 and 50 MeV/u. The experimental data revealed a significant electron cusp asymmetry, which increases towards heavier targets. This observation is inconsistent with presently available theories [6].

As a next step, the electron spectra for U^{89+}(1s^22s) ions colliding with gaseous targets of N_2 and Xe have recently been measured in the beamtime of 2016, at a projectile energy of 76 MeV/u, i.e., just above the threshold for electron impact ionization of the L-shell of uranium. In these measurements, the studied electron emission energy was extended considerably, stretching both over the full electron cusp and the binary-encounter peak. At the studied collision velocity, relativistic continuum-distorted-wave (CDW) calculations of projectile ionization show a deviation of the electron energy distribution from first-order perturbation theory due to attraction of the electron emitted from the projectile by the target nucleus. The experimental results motivate further developments of relativistic theories describing charge-changing processes in heavy-ion atom collisions.

References


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High-resolution wavelength-dispersive spectroscopy of K-shell transitions in hydrogen-like gold

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Aiming for an accurate testing of the QED effects on the ground state binding energy in high-Z, H-like ions, novel high resolution x-ray spectrometer apparatus has been developed for experiments at the Experimental Storage Ring (ESR) at GSI, Darmstadt. Namely, the twin crystal-spectrometer assembly, Bi-FOCAL, operated in the FOCUSing Compensated Asymmetric Laue geometry has been arranged for accurate x-ray spectroscopy at the ESR gas jet [1]. In a dedicated beamtime at the ESR, Lyman-α transitions of H-like Au$^{78+}$ were measured in high resolution via spectroscopy of the corresponding x rays located near 63 keV in the laboratory system [2].

This experiment represents the first high-resolution wavelength-dispersive measurement of hard x-rays stemming from a high-Z H-like ion. It demonstrates the feasibility of this method at heavy-ion storage rings, such as ESR and represents an important milestone towards achieving a sensitivity to higher-order QED effects.

Since this is a new measurement method dealing with crystal spectroscopy of relativistic high-Z ions, for obtaining an accurate result, particular attention has to be paid to systematic effects. Therefore, in the aftermath of the main experiment, few auxiliary measurements have been conducted each of the them addressing different possible sources of systematic uncertainties [3, 4, 5].

In table 1, we show a summary of the different systematic effects and the associated uncertainties on the Lyman-α1 transition energy which is used to deduce the 1s Lamb shift in H-like gold. As one can see from the table, the statistical uncertainty (stemming from the determination of the peak position) of only 2.2 eV has been achieved which is unique for a crystal spectrometer operated in the region of hard x rays of H-like high-Z ions. The systematic effects give the main contribution to the total uncertainty and have to be reduced in future runs. The ion-beam velocity can already be determined with a much higher accuracy using a high-voltage divider from the Physikalisch-Technische Bundesanstalt (PTB) in the electron-cooler terminal. With a slightly modified assembly it will also be possible to measure the gas-target position relative to the detector-crystal position in situ, which will almost entirely eliminate these systematic uncertainties avoiding supplementary experiments altogether.

References


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Table 1: Different systematic effects and the associated uncertainties on the total Lyman-α1 transition energy (preliminary results).

<table>
<thead>
<tr>
<th>Contribution</th>
<th>Value (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Preliminary Transition Energy</td>
<td>71 539.8(2.2)</td>
</tr>
<tr>
<td>Temporal Drift</td>
<td>(2.8)</td>
</tr>
<tr>
<td>Gas-Target Position</td>
<td>(13.0)</td>
</tr>
<tr>
<td>Ion-Beam Velocity</td>
<td>(4.3)</td>
</tr>
<tr>
<td>Detector-Crystal Position</td>
<td>(5.1)</td>
</tr>
</tbody>
</table>
Sympathetic cooling in two-species ion crystals at SpecTrap

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Following our previous studies of single-species ion Coulomb crystals in the SpecTrap Penning trap [1], we have investigated two-species ion crystals formed by sympathetic cooling of a second ion species injected into previously confined and laser-cooled Mg$^+$ ions from a dedicated external source [2].

The present experiments have been performed with the SpecTrap Penning trap setup [1, 3, 4], located at the HITRAP facility [5]. It uses a cylindrical, open-endcap Penning trap located in the center of a superconducting magnet for dynamic capture and confinement of externally produced ions, see figure 1. These ions can either be obtained from our dedicated pulsed source of singly charged ions [2], from other sources along the HITRAP low-energy beamline such as an electron beam ion source (EBIS), or, in future the HITRAP deceleration facility [5].

Formation of pure Mg$^+$ ion Coulomb crystals has previously been achieved by a combination of buffer gas cooling and laser cooling of externally produced Mg$^+$ ions from energies of several hundreds of eV to energies of the crystalline state below $\mu$eV within seconds [1]. We have further imaged the temporal evolution of the ion crystal structures upon injection of singly charged ions of mass-to-charge ratios $m/q = 2\ u/e$ (H$_2^+$), $m/q = 12\ u/e$ (C$^+$), $m/q = 28\ u/e$ (N$_2^+$), and $m/q = 44\ u/e$ (CO$_2^+$) into the same confinement region, see figure 2. In each case, sympathetic cooling can be observed and the results are in agreement with expectations from theory and previous experiments on centrifugal separation of ion species in a Penning trap. With ion numbers of up to $10^5$ and ion temperatures far below 1 K, these molecular ion Coulomb crystals represent ideal tools for sympathetic cooling. Such cooling is favourable when ions without suitable transitions for laser cooling are to be cooled significantly below ambient temperature, for example for precision optical spectroscopy. In particular for highly charged ions, it allows to enter the Lamb-Dicke regime in which Doppler-free spectroscopy becomes possible. The presented method and results are valuable for precision spectroscopy of medium-mass ions such as Ar$^{13+}$ and heavy ions of even higher charge states such as Bi$^{82+}$ as foreseen in the SpecTrap experiment.

![Figure 1: Sectional view of the SpecTrap Penning trap.](image1)

![Figure 2: Top: observed layer structure of the mixed-ion crystal as a function of time. Bottom: observed fluorescence signal rate as a function of time, with injection gates for CO$_2^+$ ions indicated.](image2)

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References
Proton and $\alpha$ capture studies for nuclear astrophysics at GSI storage rings


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The $^{124}$Xe(p,γ) reaction has been measured for the first time at energies around the Gamow window by using stored ions at the ESR facility. The desired beam energies below 10 MeV/u introduced new experimental challenges like windowless ions detection under UHV conditions, extremely short beam lifetimes and efficient beam deceleration and cooling, all of which have been successfully met.

In the nucleosynthesis of the so-called $p$ nuclei radiative capture reactions like (p,γ) or ($\alpha$,γ) play an important role to model the reaction network and to explain the stellar production yields in different explosive scenarios [1]. Most of the key reactions involve radioactive nuclei [2] and can be studied solely in inverse kinematics. The GSI facility offers the unique possibility to produce such exotic ions and to store them in the experimental storage ring, ESR, and eventually in the CRYRING. This setting allows one to use the limited intensities available for radioactive ions with maximum efficiency.

In a first step, fully-striped ions are stored at beam energies below 10 MeV/u. Subsequently, nuclear reactions are introduced by colliding the stored ions with the internal jet target that consists of either hydrogen or helium gas for (p,γ) or ($\alpha$,γ) reactions, respectively. Ions, which capture a proton or an α particle at the target, are separated from the stored beam in the next dipole magnet and are detected by UHV compatible double-sided silicon-strip detectors (DSSSD) with a 100% efficiency. Due to atomic interactions the lifetime of the highly charged beam is on the order of seconds. The dominant process responsible for this is the well-known radiative electron capture (REC), which can be used for cross section normalization by employing x-ray spectroscopy around the target.

The very first measurement in the ESR was performed with a beam of stable $^{124}$Xe$^{34+}$ ions decelerated to and stored at energies between 5.5 MeV/u and 8 MeV/u to study the reaction $^{124}$Xe(p,γ)$^{125}$Cs. The spatial resolution of the employed DSSSD allowed a clear identification of the (p,γ) signal sitting on a background of elastically scattered ions, as shown in Fig. 1. Similar signals could be identified for five different beam energies in the aforementioned energy range. The analysis of the data set is ongoing within a PhD project.

In the future, first reaction studies on radioactive nuclei are planned and will be carried out using the ESR setup described above. For energies below 4 MeV/u the newly installed CRYRING facility [4] is ideally suited to serve as a low-energy extension of the ESR. Corresponding experimental equipment is already being designed and will be ready for first experiments in 2018.

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A continuous data logger for the ESR current transformer

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In this work we introduce a new read-out electronics for the existing DCCT current transformer of the ESR.

Motivation

As long as coherent effects are not dominant in the beam, which is the case for beams of very high intensity, the integral power within Schottky bands are proportional to the ion beam current and hence the number of particles [1]. So a measured absolute value of current intensity is needed so that the integral power of Schottky spectra can be normalized to it. To this end usually a DC current transformer (DCCT) or alternatively a cryogenic current comparator (CCC) can be used.

This approach has two advantages: in the absence of a CCC, sensitivities out of reach to the DCCT can be realized by properly designed resonant Schottky detectors. Furthermore, using time resolved Fourier analysis it is possible to quickly follow the beam intensity within the same injection cycle. This method has been tested at the ESR storage ring at GSI using the resonant Schottky pickup [2] and the DC Current Transformer [3]. Results are available in [4].

The Hardware

The GSI DCCT electronics provides an analog signal, which after passing through a differential to serial converter in GSI operation area BG2.009, reaches the main control room. Additionally a second output was connected to a circuit based on a voltage to frequency converter, in order to make the signals available in the atomic physics data acquisition system over a 50 ohm transfer line [5]. A third low impedance output was left unused. The circuit described here has been designed to sample this output, after adaptation, using a 12-bit serial successive approximation analog to digital converter MCP3208. As the main controller, a single board computer (Raspberry Pi) running Linux (Raspbian) operating system has been utilized. The circuit is placed directly underneath the DCCT in the ESR in order to keep a short analog signal path. The digitized values are transferred over the network.

The Software

The code is written entirely in Python using a client/server structure [6]. The circuit acts as a message queue (ZeroMQ) server in the publisher/subscriber mode and broadcasts the value of the DCCT current to any subscriber available on the network. The sampling rate is set to 5 sps. Any number of clients can subscribe to the publishing server, either using the command line or the GUI and can run on any number of computers inside the network. The command line interface can be set to write out files of certain length, thereby allowing practically unlimited and continuous monitoring of ESR current over weeks of beam time. While providing a unique time stamp for every recorded sample, the server updates its clock regularly using an internet time server. The resulting data can be easily plotted or processed offline together with Schottky spectra. During the beam time of 2016, this device was successfully tested, where data were stored directly on GSI central computing cluster.

Future extensions

The electronics of the DCCT allow for certain amplification ranges. Although already implemented in the software, currently the ranges are set manually. In order to receive the ranges automatically set by the control system, an optically decoupled module has been designed in order to separate ground loops from that of the ESR. The test of this module is planned for a future beam time. The proper connection to the FAIR Control System still needs to be implemented. A similar design may be planned for experimental purposes in future storage rings of FAIR.

References


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Status of the HILITE Penning trap experiment

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The HILITE Penning trap is an ion trap developed to capture, detect and confine ions in order to provide well-defined ion-targets for laser-ion interaction studies. Therefore we apply several techniques for ion detection, ion selection and ion confinement.[1]

We have implemented the SWIFT (Stored waveform inverse Fourier transform) technique to be able to form ion targets of only one ion species. This technique has been tested at the similar ion trap ARTEMIS [2]. Based on the results of this test we have adapted our electronic circuits for noise-reduction such that we enable fast switching of the electrodes with time constants of the order of nanoseconds. These modified filters have been built and connected to the trap electrodes.

Figure 1: Picture of the assembled Penning trap - electrodes inside inner tube with applied filter boards and resonators.

For non-destructive ion detection we use resonant amplification of image currents induced in the trap electrodes by ion oscillations. To improve the sensitivity of the detection of stored ions inside the Penning trap we have built two resonators and tested them at cryogenic temperatures. To enlarge the ion spectrum, that can be detected by the resonant circuit, we will employ a varactor-diode for each resonator. We have tested the varactor-diode concerning its behaviour in the magnetic field at temperatures down to 4 K. Based on the results we have built dedicated varactor diode boards, which also support a wide tunability even at low temperatures and high magnetic fields.

In order to achieve long ion storage times the design residual gas pressure is better than $10^{-12}$ mbar. To compromise the open-endcap design of the Penning trap with a sufficient vacuum in the interaction region, a set of baffles at cryogenic temperatures is applied on each side of the trap-electrodes. As these baffles will also be used as a pulsed drift tube for ion deceleration, they are electrically isolated from the inner shield by ceramic spacers, which also support good thermal conductivity.

Figure 2: Picture of the assembled baffle system for vacuum improvement and ion deceleration.

As an ion source for the first commissioning experiments we have set up an EBIT and verified its functionality. We have produced ions inside the EBIT and have measured the extraction with a Faraday cup. The next steps will be the connection of the EBIT with our ion trap to capture ions produced inside the EBIT and check the functionality of the measurement principles of the ion trap.

Figure 3: Experimental stand with the equipped device rack.

Our recent results and the status of our experiment have been presented at the PSAS conference in Jerusalem, at the SPARC workshop in Krakow and at the MML workshop in Hamburg.

References


Status of the ARTEMIS experiment:
Purification of highly-charged ion clouds and their months-long storage

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We are currently upgrading and commissioning the ARTEMIS experiment, a Penning trap setup located at the HITRAP facility, and designed for precision microwave spectroscopy of highly charged ions \cite{1}. In combination with optical spectroscopy, this will be used to determine the magnetic moment of the electron in the presence of the extreme fields in the vicinity of the core of a highly charged ion. Within the theory of bound-state quantum electrodynamics, magnetic moments can be calculated to high accuracies. With the foreseen measurements at relative accuracies on the ppb level and beyond, it is possible to test such calculations with high stringency. The method of choice is the so-called laser-microwave double-resonance spectroscopy, utilizing the fact that for medium-heavy few-electron ions the fine structure, and for some heavy highly-charged ions the hyperfine-structure splitting is in the optical regime. These ions, such as \textsuperscript{207}Pb\textsuperscript{81+} and \textsuperscript{209}Bi\textsuperscript{82+}, are foreseen to be available within the framework of the HITRAP facility. For efficient light collection, the Penning trap features a dedicated half-open design \cite{2}.

For first off-line tests, the \textsuperscript{40}Ar\textsuperscript{13+} ion has been chosen. It has a spinless nucleus, so that the magnetic moment (g-factor) of the 2p-electron can be measured. Due to the high magnetic field of 7 T also first laboratory measurements of higher-order Zeeman effects can be performed \cite{2}. To that end, we have successfully and routinely operated an in-trap source of highly charged ions, similar to a miniature electron beam ion trap, and have created ions up to \textsuperscript{Ar}\textsuperscript{16+} and \textsuperscript{W}\textsuperscript{26+}.

![Figure 1](image1.png)

Figure 1: Detected argon ion charge state spectrum for different storage times. The inset indicates small changes, used for the residual gas pressure measurements.

We are able to store these ions over the course of many days, detect them non-destructively by use of dedicated radio-frequency resonators \cite{4}, and to cool their motion. We have used the cooled ion cloud to perform a residual gas pressure determination in our trap chamber. With a measured half-life of 78 days, an upper limit of the residual gas pressure of 9 \times 10^-16 mbar has been estimated. Such a measurement is depicted in figure 1.

![Figure 2](image2.png)

Figure 2: The SWIFT cleaning technique performed on an ion cloud in the creation trap of ARTEMIS.

We have demonstrated preparation of pure ion clouds by application of the Stored Waveform Inverse Fourier Transform technique (SWIFT). It enables selective excitation of different charge states in order to remove unwanted ion species from the trap. Figure 2 demonstrates the procedure required to achieve a pure cloud of \textsuperscript{Ar}\textsuperscript{13+} from a rather extreme example of an ion cloud in our trap.

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\begin{enumerate}
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Progress of experimental systems for CRYRING@ESR∗†

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CRYRING@ESR is a new heavy ion storage ring facility at GSI/FAIR and is presently under construction [1-2]. The former Swedish CRYRING was modernized and adapted to its new location. After almost two years, reassembly of the ring in general has been completed and vacuum pumping could be started. Also, the beam transport from ESR to CRYRING has been completed. Figure 1 shows a photograph of the injector beamline and the ring at the end of 2016. During the course of 2016, already the first two beamtime campaigns were dedicated to the commissioning of CRYRING@ESR. During these periods, the low-energy beam extraction from ESR and transport towards CRYRING, as well as — using a beam produced in the local injector of CRYRING — the “first turn” of ions in the ring could be successfully demonstrated. More details about the commissioning are given in [3]. For the machine, the directions are set for going from first turn to a long-lived stored beam. Thus, next steps will be establishing the required ultrahigh vacuum conditions for a long ion beam lifetime, as well as electron cooler operation.

With the machine gradually progressing into regular operation, the experimental systems are presently also being prepared, and CRYRING@ESR in fact offers exciting research opportunities for research on highly charged ions for a large range of scientific fields. In the future, all ions presently available from the GSI accelerator chain can be transported and stored at low energies, between ~ 0.05 to 15 MeV/u with ion beam lifetimes between few seconds to ~ 15 minutes, depending on charge state and energy. Hence, SPARC and FLAIR, but also NuSTAR have formulated extensive research programmes for this facility [4]. The low energy conditions will allow for precision spectroscopy and thus, e.g., allow one to test non-perturbative strong field QED, or to study transient quasi-molecular systems. Further, at the border between atomic and nuclear physics, nuclear size effects, hyperfine interactions or exotic couplings between the electronic shell and the nucleus of an ion may be explored. In nuclear physics, storage of bare nuclei at low energies permits to determine fragment distributions unmasked by atomic physics. Under these conditions, e.g., nuclear reactions at the Coulomb barrier or nucleosynthesis of heavy elements in the Gamow window of the p-process may be analyzed.

A first generation of experiments at CRYRING@ESR

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References

SPARC experiments with highly charged ions at the HESR of FAIR

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Introduction

One of the aims of the SPARC collaboration [1] at FAIR is to perform precision atomic physics experiments with highly charged heavy ions at the High Energy Storage Ring (HESR).

An internal target is indispensably an integral part for many such experiments. Ions with different charge states, which are obtained as a result of interaction of an ion beam with the target, need to be effectively separated and detected. In this work we present ion optical studies unambiguously showing the feasibility of SPARC experiments at the HESR.

Target location

The SPARC collaboration at FAIR [1] aims at atomic physics research with highly charged ions (HCl) in energy domains previously not accessible for precision experiments [2,3]. Here, the High Energy Storage Ring (HESR) will allow for storing stochastically and/or electron cooled HCIs up to energies of ~5 A GeV.

The missing dipole concept in the HESR enables an installation of internal target stations [4] in two arcs (ROI 1 and 2) [5] (see Figure 1).

However, only one dipole magnet is then available for charge state separation [5].

Figure 2 shows a separation of the primary fully ionized \(^{238}\text{U}^{92+}\) uranium beam (2-sigma emittance) and two charge states (\(^{238}\text{U}^{91+}\), \(^{238}\text{U}^{90+}\)) obtained after the reaction in the target (ROI 2). The ion optical computations prove that the resolution of the charge states at the particle detector, which is placed 3 m after the last defocusing quadrupole, is more than sufficient. For experiments with lighter beams, the separation improves further since \(\Delta Q/Q\) increases [5]. As a conclusion, the experiments with internal target, placed in the missing dipole gap in ROI 2, are feasible [7]. Examples of the growing number of the proposed precision experiments in the HESR, which will thus be enabled, can be found in [8-14].

References


Figure 1: Two target locations in the HESR and the corresponding regions of interest (ROI) can be seen.

The experimental conditions in the ROI 1 region, which was initially proposed for the SPARC target location [6], have a complication: ions, recombined in the electron cooler and the rest gas along the long straight section, will produce an unwanted background. This situation is avoided in the ROI 2 case resulting in much cleaner experimental conditions.
Ion trajectory simulations for the purification system of the FISIC experimental program

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With the Fast Ion-Slow Ion collisions (FISIC) experimental program [1], we propose an experimental crossed-beam arrangement in the so-called intermediate regime (the regime in which the ion stopping power is maximum) for a wide range of projectile-target combinations with an ultimate control of experimental conditions to measure absolute cross-sections of fundamental atomic processes. Up to now, only a few crossed-ion-beam experiments have been performed for light ions in the low energy-energy domain [2].

Simulation results

Crossing two multi-charged ion beams, under well-controlled conditions, has always been a very challenging task, whatever the physics under consideration. For such a challenging experimental project, many technical barriers have to be overcome. For the low-energy beam line, one of them is to prevent pollution mainly due to electron capture from the residual gas before the collision point. To control the charge state of keV/u ions, a new Omega type purification system has been developed. This system consists of four cylindrical deflectors with Matsuda plates allowing us to generate toroidal electric fields. This particular arrangement allows to get the exit beam back on the initial beam axis. Ion trajectory simulations have been performed with the SIMION 8.1 version software in order to optimize the geometry of the purification system. To test different configurations, we have used the batch mode operation of SIMION with a Lua code. In the simulations, we generate an ion beam with an energy of 20 keV/q and a large emittance of 60 π mm.mrad with a Gaussian spatial distribution ($\sigma_x = 0$, $\sigma_y = 2$mm, x being the ion propagation axis). To test the purification system, we generate ions having the same mass but with different charge states. The voltages on the cylindrical deflectors are set in such a way that only the desired charge state is allowed to pass through the purification system while the other charge states are stopped. As an example, simulated ion trajectories for Ar$^{15+}$ with q = 15, 16, 17 and 18 are shown in Fig. 1 with voltages that permit only the transmission of Ar$^{17+}$ ions. Therefore, a clean Ar$^{17+}$ ion beam is obtained just before the collision with the fast ion beam. Simulations for different ion species such as C$^{6+}$, N$^{16+}$, O$^{17+}$, and Ne$^{20+}$ have also been carried out with success showing the versatility of this purification system.

Figure 1: Ion trajectory simulations in the omega-type purification system. The voltages are set to permit only the transmission of Ar$^{17+}$ ions.

Future work

A dedicated chamber for the purification system has been designed and is under construction. Experimental tests are scheduled first at the ARIBE facility at Caen. Meanwhile, simulations of the collision zone together with the post-collision electrostatic charge state analyser of the low-energy ions are under progress.

References


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Parallel plasma description of ions stored in Penning traps∗

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The HITRAP (Highly Charged Ions Trap) facility at the GSI allows to investigate slow highly charged ions up to U 92+. The most important part of the facility is the Penning trap, which allows the trapping of charged ions.

A microscopic simulation of the ion cloud is only possible to a limited extent. Because of the computational effort of $O(N^2)$ a system of $10^5$ ions, as occurs in the ion trap, leads to long simulation times, which makes long-term simulations impossible. Even if we parallelize the problem and have access to an infinite number of resources, a simulation would take about 15 years [1].

Plasma description

The macroscopic description provides further access [2]. The ion-ion interaction is avoided and we look at the system of ions as a continuum similar to a charged liquid. We use a statistical approach in which the positions and velocities of the ions are given by a probability. This means that the dynamics of the ion cloud is described by the dynamics of the density $f(r,v,t)$ in the phase space, which describes the probability of finding an ion with a given velocity at a certain point in space.

The dynamics of the single particle density $f(r,v,t)$ is given by the Vlasov equation

$$\frac{\partial f}{\partial t} + v \cdot \frac{\partial f}{\partial r} + \frac{F_{\text{ext}}}{m} \cdot \frac{\partial f}{\partial v} = 0 \quad (1)$$

which is an exact description of non-interacting particles. The force $F$ is given by the external electromagnetic fields. The interaction of the ions among each other is described by an mean-field $\Phi(r,t)$. This is given by the charge density $\rho = q \int f d^3v$ which is given by the ion density, where $\Phi$ is the solution of the Poisson’s equilibrium $\Delta \Phi \epsilon = \rho$. Together with the associated force $-q \nabla \phi$ and the Vlasov equation (1), we obtain the non-linear Vlasov-Poisson equations

$$\frac{\partial f}{\partial t} + v \cdot \frac{\partial f}{\partial r} = -\frac{q}{m} (-\nabla \phi + F_{\text{ext}}) \cdot \frac{\partial f}{\partial v} \quad (2)$$

Parallelisation

The Vlasov-Poisson equations (2) have to be solved numerically for practical applications. A grid-based hybrid MPI/OpenCL solution called TRAPSim was implemented to run the simulations on the GSI Green IT Cube in order to minimize the computational effort.

The L-CSC cluster consists of 160 compute nodes, each with four AMD FirePro S9150 GPUs, two CPUs and 256 GB memory.

The implementation was tested with a system of $2^{17}$ ions and a grid of $64 \times 64 \times 128$ points. In Table 1 we see the measurement data of the runtime for an iteration. $g$ is the number of GPUs and $t$ the runtime in seconds. $s$ the speedup, $f$ the serial fraction and $e$ the efficiency, as defined in [3].

<table>
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<td>-</td>
<td>0.97</td>
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Table 1: Measuring of the parallel GPU Performance.

We see that the serial fraction $f$ decreases. The reason for this is that by increasing the number of GPUs we get more cache and memory bandwidth that reduces the overhead.

Which maximum speed-up can be expected? Due to Amdahl’s law [4] we can calculate the maximum speed-up with the limit

$$\lim_{g \to +\infty} s(g) = \frac{1}{f} \quad (3)$$

If we choose for $f = 4.2E-3$, the limit is $s_{\infty} \approx 238$. The runtime of the simulation depends strongly on the selected grid. A detailed analysis is needed to find the minimal grid that still describes physics.

References


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Backscattered electron emission from gold nanoparticle after protons impact: Experiments and simulations*

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It has been shown that the presence of gold nanoparticles (GNPs) can increase cell damage during radiotherapy treatment, possibly due to emission of low energy electron around GNPs. Reproducing the interaction between protons beam and GNPs by Monte Carlo simulation requires cross section data which are still incomplete. In this context, theoretical models need experimental validation. This work aims at a comparison of simulation results calculated with the TRAX code and with experimental data obtained from irradiation of GNPs deposited on Carbon thin film.

The TRAX code is dedicated to the description of low-energy electron emission and transport in solids after proton irradiation [1]. Sets of interaction cross sections for electron with energies down to 1 eV are available for different materials and allows us to reproduce ionization, excitation and elastic scattering. Standard elastic scattering cross sections can be calculated using the screened Rutherford approximation or the Partial Wave Analysis method, respectively, for low and high Z. Ionization induced by electron is taken into account by applying the binary encounter Bethe model. Auger electron emission probabilities are implemented in the code using Livermore Evaluated Atomic Data Library.

Experimental data were gathered at the University of Namur. GNPs were deposited on thin Carbon films using the magnetron sputtering method developed in Namur [2]. Gold nanoparticles size and distribution were observed with transmitted electron microscopy (see Figure 1). The experimental setup dedicated to the secondary electron emission is described in our paper published in Nuclear Instruments and Methods in Physics Research Section B [3]. This paper presented secondary electron emission result obtained from carbon thin film (50 nm and 100 nm thick) and gold thin film (200 nm thick).

Electron energy spectra resulting from 2 MeV proton bombardment on carbon and gold nanoparticles deposited on carbon substrate are displayed on Figure 2. Preliminary TRAX simulation of GNPs deposited on carbon are also presented. As expected, presence of GNPs increase the secondary electron yield. It turns out that the simulation is quite similar to the experimental results at low energy (< 270 eV). The Auger peak at ~270 eV appears more pronounced in TRAX simulation. The difference in electron yield above 270 eV has to be studied. As it is a preliminary simulation, this could be due to geometrical errors. This will be discussed in further works.

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References
Test of a new silicon microcalorimeter array at the ESR

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Silicon microcalorimeters for high-precision X-ray spectroscopy have been developed at GSI and the University of Mainz since more than two decades [1]. These microcalorimeters are based on arrays of silicon thermometers [2] and X-ray absorbers of lead or tin [3] to obtain high quantum efficiency for X-ray energies of around 100 keV. With such detectors, an excellent energy resolution of $\Delta E_{FWHM} = 40 – 60$ eV for a X-ray energy of 60 keV has been demonstrated [3, 4]. Two detector arrays were applied in two successful experiments at the ESR for the determination of the 1s Lamb Shift on hydrogen-like lead and gold ions [4].

For experiments at FAIR, a new detector array with a larger detector solid angle is currently developed within the SPARC collaboration. As microcalorimeters detect the energy of a photon as a heat signal, low operating temperatures of below 100 mK are mandatory to obtain high energy resolution. Accordingly, these detectors are operated in a $^3$He/$^4$He dilution refrigerator. In the past, these detectors were operated in a cryostat with a specially adapted side-arm which fits to the gas-jet target geometry at the ESR and allows bringing the detectors as close to the interaction region of ion beam and gas jet as possible [3]. This cryostat has been used in the above-mentioned Lamb Shift experiments and performed well at the ESR, but the use of liquid helium and nitrogen as coolants makes its application cost-intensive. In addition, interruptions of beamtime for refilling the cryogenics are unavoidable. To overcome this disadvantage, a new dry cryostat, which is equipped with a pulse-tube cooler to reach 4 K and a $^3$He/$^4$He dilution stage, was designed in cooperation with the company BlueFors and commissioned in Giessen in the last three years [5]. In spring 2016, this new cryostat was for the first time set up at the ESR gas-jet target to test its performance.

The test run was performed at the ESR in combination with several other detector systems which are developed within SPARC. Our detector system was set up at the 145° port of the ESR gas-jet target, similar to the setup of the Lamb Shift experiments [4]. The cryostat performed flawlessly at the ESR over six weeks, during which several cooldown cycles were completed. The quick cooling and warming up as well as the remotely controlled operation provided a great advantage because several adjustments of the setup were found to be necessary.

In the test experiment, X-rays were detected which had been produced by interaction of an ion beam with a gas-jet target. This experimental setup has been described in detail in many publications, i.e. in [4]. A beam of bare or highly-charged ions is injected into the ESR, stored and cooled. After the cooling cycle is finished, a gas-jet, which is oriented perpendicular to the ion beam, is switched on.

Figure 1: The upper panel displays a photograph of the new, more compact detector which was tested in 2016 at the ESR in the first campaign with uranium ions. The lower panel shows the actual microcalorimeter array without its protective aluminum cover.
Figure 2: Left: X-ray spectrum from the interaction of a 30.85 MeV/u ($\beta \approx 25.1\%$) $^{124}$Xe$^{54+}$ beam with a N$_2$ target, not corrected for the Doppler shift. The displayed spectrum was measured with one microcalorimeter pixel within 12.5 hours. The Xe-K$_{\alpha 1,2}$, K$_{\beta}$ and K-REC could be assigned. The insert shows the region of the K lines in an amplified scale. Right: X-ray spectrum from the interaction of a 5.95 MeV/u ($\beta \approx 11.2\%$) $^{124}$Xe$^{54+}$ beam with a H$_2$ target, not corrected for the Doppler shift. The displayed spectrum is the sum of six microcalorimeter spectra which were recorded within 22 hours. The Xe-K$_{\alpha 1,2}$ can be identified in the spectrum. The K-REC is not visible because the probability for this process is low at low ion energies. The insert shows the region of the K lines in an amplified scale. These spectra were obtained with the old detector array described in [4] which was mounted in the new dry dilution refrigerator for the second measurement campaign.

While the ions cross the gas-jet, part of them may capture an electron and emit X-rays, which are detected by our microcalorimeter. The test experiment consisted of two campaigns: The first campaign provided a beam of helium-like uranium which interacted with a gas-jet of molecular nitrogen. Accordingly, X-rays from lithium-like uranium were detected. This was not optimal for our detector, because the X-ray energies were of the order of 15 keV or below, with the most prominent line at 4.5 keV. Our setup is optimized for energies around 100 keV. Accordingly, our sensitivity for these low X-ray energies is rather small. X-ray energies below 10 keV cannot pass the current windows of our cryostat. Nonetheless, we could distinguish X-rays coming from the interaction of ion beam and gas-jet, but the energy resolution was unexpectedly low. It was found that the observed signal amplitude was considerably smaller than expected. The reasons for this behavior are currently under investigation.

To investigate if the low energy resolution was only due to the low signal-to-noise ratio or if other aspects contributed as well, in the break between first and second campaign we exchanged the new detector with the old detector which had been used in former experiments [4]. In the second campaign, hydrogen-like xenon ions interacted with a gas-jet of hydrogen. This configuration in combination with a very low beam energy resulted in very small count rates, which in turn caused us to remove the calibration sources that we usually observe in parallel to the actual experiments in order to have a permanent energy calibration. This removal was possible because the cryostat maintained a very stable operation temperature over all times.

Example spectra for two different ion energies are displayed in figure 2. Despite the low count rates, we observed Lyman-$\alpha$ transitions from hydrogen-like xenon with energies around 30 keV as well as X-rays from direct radiative recombination into the K shell (K-REC). The energy resolution obtained was around 200 eV, which is comparable to the energy resolution obtained in past experiments [4]. Optimization and further data analysis is currently in progress. In parallel, the next-generation microcalorimeter array with 96 pixels and an active area of approximately 1 cm$^2$ is in preparation. We expect that this larger array with the designed performance will be available for experiments in 2018.

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References


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Recent developments for the CRYRING@ESR transverse electron target∗

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As part of the instrumentation of the upcoming storage ring CRYRING@ESR a ribbon-shaped free-electron target for atomic physics electron-ion collision studies is presently being developed and constructed [1, 2, 3, 4]. The target operates in crossed-beams collision geometry, i.e., with an interaction angle of 90° with respect to the ion beam circulating in the ring.

Within the last year, simulations for the electrode configuration were carried out. The according design is based on a multi-electrode electron gun [5, 6] originally developed for the Giessen low-energy single-pass electron-ion collision facility [7]. Yet, the present electrode layout has been substantially altered in order to adapt the electron target to the experimental environment at CRYRING@ESR. The electron gun has several advantages, among others: (i) the possibility to set the electrode potentials to a large extent independently from each other, yielding a higher flexibility in operation modes, (ii) a very homogeneous electron density (ne) in the interaction region, (iii) minimization of angular misalignments with respect to the ion beam, (iv) clearing electrodes in order to avoid trapping of slow ions from ionization of residual gas, (v) a new decelerating collector with lower heat dissipation, (vi) a large interaction gap to provide space for the circulating ion beam, and, (vii) the realization of a high density mode with ne > 1 · 10⁹ cm⁻³.

In addition, the major features of the mechanical and vacuum layout have been worked-out (Fig. 1). The electron target can be fully retracted behind a gate valve and, thus, can be separated from the ring vacuum. This allows maintenance work to be carried out without breaking of the ring vacuum. A turbo pump (Pfeiffer HIPACE 300H) with high compression for light gases and a cryo pump will be used to fulfill the stringent vacuum requirements (p ≈ 10⁻¹¹ mbar) of CRYRING@ESR. Two large viewing ports will provide spectroscopic access to the interaction region.

The experimental electron target station has been optimized to enable a joint installation with the gas-jet target in the experimental section YR09 (Fig. 1). The concurrent setup allows for a more flexible usage of test and experimental beam times since no vacuum-breaking changes of setups needs to be performed. Additional synergies such as for diagnosis or instrumentation or even joint experiments are currently being investigated. At present, still a few details and short-noticed changes need to be clarified. It is planned that an off-line version of the target will be available in the first half of 2018, and, depending on additional funding, the target will be available in CRYRING@ESR in 2021.

References

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Dynamic compression of diffractograms for an improved convergence in ptychography

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The quality of ptychographic reconstructions mainly depends on the signal redundancy of the diffractograms. We present a method for improving diffraction images used for ptychography to enhance the convergence time and object visibility.

Dynamic compression of redundant fringes

Ptychography uses redundant data from overlapping illumination spots to overcome the isolation constraint known from classical CDI. The amount of redundant information in the diffractograms gives a criterion for quality of reconstruction and time of convergence [1].

Figure 1: Different ptychographic scan positions (left and right) of an object (top) lead to varying diffractograms (bottom). The overlapping area of these two scan points (middle row) leads to a diffractogram, which consists of redundant fringes.

The usage of a Gaussian intensity profile for the illumination spot leads to lower intensities of the redundant illuminated parts of the object. Therefore the intensity of the supernumerous fringes in the fourier space decreases. We use a method called dynamic compression (DC) or tone mapping to amplify these weak information in the diffractogram. Due to the ambiguity of the diffraction pattern, it is difficult to selectively identify and amplify the redundant pattern. Because of the low intensity in the overlapping area, it is more probable to find redundant information in the weak fringes. Thus weak signals of the diffractograms getting increased with respect to high intensity fringes under usage of an adapted local gradient tone mapping operator [2] (fig. 2).

Figure 2: Dynamic compression (right) of a diffractogram (left) with background subtraction.

Ptychographic reconstruction

The ptychographic reconstruction with the ePIE-algorithm [3] of the object from fig.1 is shown in fig.3. For both reconstructions the number of iterations was fixed to 10. Whereas the object without dynamic compressed signals are barely visible, the algorithm using enhanced diffractograms was able to converge to a solution, which is more similar to the scanned object.

Figure 3: Reconstruction of the object shown in fig.1 without DC (left) and with DC (right).

References

Online monitoring of XUV spectra from high-harmonic generation by surface reflectivity measurements with particle detectors


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We present reflection measurements of cesium iodide (CsI)-coated and uncoated steel surfaces with shaped XUV pulses. We measure the spectrally integrated signal from both surfaces and show that the ratio of the two measured signals is a sensitive probe of changes in the incident XUV spectrum. Additionally, a disagreement between the measured ratio and the calculated number can be explained by a leakage of IR radiation through one of the metal filters. This simple setup allows for the monitoring of the IR leakage and spectral stability simultaneously. The benefits of our approach are a spectrally sensitive diagnosis of the XUV radiation at the interaction place of time-resolved XUV experiments and the detection of infrared leak light though metal filters in high-harmonic generation (HHG) experiments. Our obtained results are of interest for time-resolved XUV experiments presenting an additional diagnostic directly in the interaction region and for small footprint XUV beamline diagnostics. For the experiments we used spectrally shaped XUV pulses from high-harmonic generation in an argon-filled capillary (Fig. 1, detailed experimental description in [1] and [2]). The XUV radiation is either directly launched into a XUV spectrometer or detected with a Channeltron after reflection from an uncoated or CsI-coated stainless steel plate. The polished surfaces were coated by a vacuum evaporation process with a 300-nm-thick layer of high purity CsI [3]. The secondary electron signal from the Channeltron is amplified, discriminated and measured by a multiscaler with sub-nanosecond resolution. From the XUV spectra (Fig. 2a) the reflectivity ratio between the two surfaces can be calculated using tabulated values [4]. A comparison with reflection measurements using a Channeltron shows agreement within less than 3% difference to the calculation (Fig. 2b), but in case of residual IR light a stronger deviation is visible (≈10%).

Further investigations with high-harmonics with residual IR light showed, that the Channeltron signal is sensitive to femtosecond laser light in an XUV-beam, although the Channeltron is regarded as solar blind. In the temporally resolved secondary electron signal from the Channeltron the interaction with femtosecond IR pulses can be clearly attributed to certain time windows [1].

References

YAP:Ce-based scintillation devices for heavy ion detection within the Fit-FISIC project∗

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With the advancing realization of the novel FAIR accelerator and ion storage complex, in particular the impending commencement of CRYRING operations (FAIR Phase 0), planned experiments such as the Franco-German Fit-FISIC project (First steps towards atomic physics of Fast Ion–Slow Ion Collisions, [1,2]) depend on the availability of robust ion detectors. Among the quite extensive variety of established detection methods, the use of scintillation detectors provides a solution that is both economical as well as versatile with respect to the ion energies and species accessible [3]. While common plastic scintillators usually suffer from fatal radiation damage due to the localized nature of ion energy deposition – the so-called Bragg peak –, crystalline substances such as YAP:Ce (cerium-doped yttrium aluminium perovskite) have been known to exhibit a significant degree of radiation hardness [4].

Commonly the readout of such scintillator detectors is accomplished with a photomultiplier tube of suitable spectral sensitivity. However, some experiments require position resolution that this approach fails to provide, e.g. to distinguish the trajectories taken by different charge states. A promising alternative is the use of so-called silicon photomultiplier (SiPM) devices. This novel detector type, sometimes called “multi-pixel photon counters”, can be described as an array of parallelly connected avalanche photodiodes with a typical size of 25 μm, and provides a compact, low-voltage and easily tileable readout solution. In addition, the devices’ inherent high gain affords single-particle detection efficiency.

Pursuing this approach, a demonstrator setup is presently investigated at Helmholtz Institute Jena. It consists of an 8 × 8 assembly of square SiPM devices with an edge length of 6 mm each, manufactured by SensL Technologies Ltd., and readout electronics furnished by Vertilon Corporation. A dedicated breakout board allows for custom signal processing, e.g. for use in experiments where timing information is critical. The actual YAP:Ce scintillator crystal has a thickness of 1 mm and was manufactured by CRY-TUR spol.s.r.o. It is attached to the SiPM array with an aluminium frame for easy mounting; a photograph of this detector head is reproduced as an inset in figure 1. At the scintillator’s primary emission wavelength of 370 μm, the photon detection efficiency of the SiPMs is on the order of a favorable 40%, on par with typical values achieved with conventional photomultiplier tubes.

Figure 1: Oscilloscope signal from the SiPM readout of a 1 mm-thickness YAP:Ce crystal scintillator, irradiated with 662 keV photons from a 137Cs source. The inset shows the scintillator array bonded to the SiPM detector head, of which only the 8 × 8 segmentation is visible.

Figure 1 also displays the oscilloscope trace of the observed signal when the scintillator–SiPM assembly is irradiated with 662 keV photons from a 137Cs γ source. The peak exhibits a steep rising flank with a rise time of approximately 50 ns (10%–90% of maximum signal). The falling flank, on the other hand, is decidedly longer, with its slope primarily determined by the quenching resistors that halt the avalanche. In any case, operation at a maximum event rate of some 100 kHz is obviously feasible.

Pending availability of the necessary vacuum components, a characterization measurement with ion energies of ca. 10 MeV is currently being set up at HI Jena.

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Status report on a Si(Li)-Compton polarimeter of SPARC - 3D-readout of a thick double-sided Si(Li) strip detector

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The x-ray polarization and spectroscopy research program of the SPARC collaboration [1] at FAIR and GSI depends strongly on the performance of the available Compton polarimeter instruments within the community. During the last years we applied our Si(Li)- and Ge(i)-Compton polarimeters in several accelerator-based beam time campaigns [2,3] at GSI, DESY, ESRF, etc.. A consequent development of the detector and readout techniques led to the design and realization of our first Si(Li)-DSSD (double-sided strip detector with 1.0mm² spatial resolution) with 64 preamplifiers with a 1st-stage at cryogenic temperature. This change in readout technique improves the energy resolution to 850eV-900eV at 60keV (ground-side) and 1200eV at 60keV (HV-side) compared to 2000eV at 60keV with a readout with all parts (except the DSSD) kept at room-temperature. The improved energy resolution decreased the lower energy threshold for a reliable event reconstruction with respect to linear polarization detection of incident photons from 70keV down to 40keV. With this progress we are now able to study a huge variety of transitions in atomic systems that were not accessible for us up to now. Recently we studied, with a focus on future experiments at FAIR higher event complexities and rates, the possibilities of a reliable identification of the point of interaction within the detector crystal (z-component). The relatively simple geometry of the DSSD allows for identification of the x- and y- position by superposition of the front side and back side strips of the detector. The z-position may be measured by the time of arrival of the electron cloud (hole cloud respectively) at the strip contacts. For our 9.0mm thick Si(Li)-DSSD we measured an electron drift velocity of 37μm/ns in the depleted bulk. This result agrees well with the values known from literature. From the edges of the drift time spectrum one can derive a time resolution of 20-30ns for the measurement of the z-component. This corresponds to a measurement with a collimated photon fan beam (approx. 1mm FWHM width at the detector surface) hitting the detector at an angle of 35 degree with respect to the surface (figure 1). With the knowledge of the z-component the quality of polarization measurements can be improved with the respect to background suppression. The same is true if we turn the argument of the z-component around. The typical time coincidence resolution with a fast (typically 10ns resolution) external signal is in the order of 80-120ns due to the different path length of the charge depending on the location of the point of interaction inside the bulk. With the z-information this uncertainty in time can be reduced significantly resulting in an improved event identification efficiency.

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Commissioning of a detection system for forward emitted XUV photons


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The Institut für Kernphysik in Münster has developed a system for in-vacuum detection of forward emitted fluorescence photons created in laser-spectroscopy experiments with highly-charged ions at the experimental storage ring (ESR) [1]. The detector is optimized for XUV photons in a wavelength region around 10 nm. It will be used in an anti-collinear laser-spectroscopy measurement of the 3P0 – 3P1 splitting in beryllium-like krypton [2]. Since ions in the ESR are stored at relativistic velocities, the emission of fluorescence photons is forward peaked due to the Lorentz boost. Therefore, the detector consists of a movable cathode plate with a central slit that can be positioned around the ion beam axis collecting mainly forward emitted photons (figure 1, left). Secondary electrons emitted from the CsI-coated cathode are guided via electric and magnetic fields provided by two solenoid coils and a system of ring electrodes to a multi-channelplate detector placed inside the ultra-high vacuum of the ESR (10−11 mbar).

Figure 1: Left: CAD drawing of the XUV detector setup at the ESR. Right: Schematic of the laser spectroscopy setup for the test beam time conducted in summer 2016.

For tests of the pulsed laser system for laser cooling, and for commissioning of the XUV detection system, three days of ESR beam time with 12C3+ ions could be used in summer 2016. In an anti-collinear laser-spectroscopy setup (figure 1, right) the 2s1/2 – 2p1/2 and 2s1/2 – 2p3/2 transitions at λ ≈ 155 nm were investigated. The ions were stored at β ≈ 0.47 which results in a fluorescence wavelength Doppler shifted to approx. 93 nm. To excite the transitions, two laser systems were available: a pulsed laser system from HZDR/TU-Dresden at 257 nm [3], and a tunable cw laser system from TU-Darmstadt, also at 257 nm [4]. Each transition was measured by varying the electron cooler voltage, thus changing the velocity of the stored ions inside the ESR (example measurement shown in figure 2, left). As can be seen from the MCP signal, the fluorescence is clearly distinguishable from the background, despite the detector not being optimized for a wavelength region around 93 nm. The maximum in the fluorescence yield, occurring when the ions are in resonance with the laser, was determined for different values of the electron cooler current. This way, space charge effects shifting the ion velocity, caused by the intense electron beam of the e-cooler, could be investigated systematically. A preliminary result for the 2s1/2 – 2p3/2-transition is shown in figure 2, right. In conclusion, the XUV detection system was successfully commissioned and both fine structure transitions in 12C3+ ions could be measured. The XUV detector was found to be very sensitive to the fluorescence from the ions, but also susceptible to background coming from the ion beam and laser stray light. Therefore, background reduction methods are being investigated for a detector upgrade in prospect of a possible ESR beam time in 2018 or 2019.

References

In July 2016, during a test beamtime of 3 days, test experiments concerning laser spectroscopy and laser cooling of Li-like carbon ion beams could be performed at the ESR. The new XUV detector system (Münster university) for fluorescence measurements could be tested, as well as the new pulsed laser system (HZDR/TU Dresden) and the cw laser system (TU Darmstadt) for laser cooling of the stored and bunched ion beams. Both laser systems have been used for laser spectroscopy and laser cooling. A high-voltage divider (TU Darmstadt) was used to record the voltage of the electron cooler during laser spectroscopy measurements.

The laser system from the TU Darmstadt is a fast and broad scanning cw-laser system [1]. The system from the HZDR/TU Dresden is a pulsed laser system with a repetition rate up to 1 MHz, delivering short pulses (∼ps) [2]. The group in Münster has designed and built an XUV detector for the ESR [3]. A pure beam of up to 1 mA of carbon ions was injected into the ESR at an energy of 122 MeV/u. The $2s_{1/2} - 2p_{1/2,3/2}$ transitions in these Li-like carbon ions have a wavelength around 155 nm and can, due to the Doppler boost at the ion velocity $\beta = v/c = 0.47$, be excited by the 257 nm laser systems.

During the first part of the test beamtime, we focussed on tests of the XUV detector system. Firstly, the system was able to move in and out of the beam without disturbing it. Secondly, the voltages and magnetic field were applied and the fluorescence from the laser-excited ions was seen. Thirdly, the resonance (=transition frequency) was sought by scanning the maximum in the fluorescence whilst scanning the high-voltage from the ESR electron cooler in the smallest steps over the expected value. A typical result, for a fixed value of the electron cooler current, is shown in figure 1. Here, time is plotted vertically, the (189th harmonic of the) ion revolution frequency horizontally, and the Schottky power density of the signal in color. This scan was performed for many different values of the electron cooler current to account for space charge effects (see [3]). This way, both transitions in carbon have been determined precisely. The data analysis is still ongoing.

The second part of the test beamtime concentrated on laser cooling using the cw laser, and especially the pulsed laser. The ion beam was moderately bunched (10-15 bunches) by the exciter. In figure 2, using the cw laser, it can be seen that the ion velocity changes as the bunching frequency is being varied. At the optimal point, the many sidebands around the central line (red) should vanish, indicating that the amplitudes of the synchrotron oscillations are very small. In contrast to the beamtime in 2012, no laser cooling could be observed in 2016, mainly due to time contraints. The data is still under analysis. However, for the first time, a clear interaction of the ion beam and the pulsed laser has been observed. This important result motivates further studies with pulsed laser systems.

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Recent Work on the Darmstadt laser systems for laser cooling of relativistic ion beams at SIS100∗

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We are developing two laser systems for laser cooling of highly relativistic ion beams at the SIS100. The first is a continuous wave system with fast and broad tuning capabilities, the second is a pulsed system with variable pulse duration and a high repetition rate. In this contribution we discuss recent progress in the development of these systems.

Laser cooling of relativistic ion beams was successfully demonstrated at the ESR during beam times in 2004, 2006 and 2012 [1]. With this technique a relative longitudinal momentum spread of less than \( \Delta p/p = 10^{-6} \) was achieved [2]. It was shown that intra beam scattering is a potential source of heating [3]. Simultaneous cooling with cw and pulsed lasers is a possible solution to overcome this limitation and will be implemented as the sole source of beam cooling at the SIS100 of FAIR.

The cw laser system was described in detail in a recent publication [6]. In brief, we use the radiation from an external cavity diode laser (ECDL), amplify it in a fiber amplifier and then use two build-up cavities in order to convert the IR radiation into the UV range using LBO and BBO crystals, respectively. A second ECDL locked to a wavemeter is employed to off-set lock the first and guarantee smooth and fast tuning. While the performance of the system during the beamtimes in 2012 and 2016 was very reliable, the system employs several locking and control circuits in order to operate. Therefore, a goal has been in our recent work to simplify the setup.

In light of this goal we have improved upon the performance of the fiber amplifier and replaced the first build-up cavity by a single pass configuration using a periodically poled LiNbO\(_3\) crystal (PPLN). Overall efficiency was lower than in the previous setup. However, we managed to generate up to 2.4 W of green radiation at 514 nm compared to 5 W in the previous setup. We need to reduce the green power entering the second cavity in order not to exceed the damage threshold of the BBO crystal. The tuning capabilities are slightly narrower due to the limited temperature acceptance of PPLN, but for a combined cw-pulsed laser cooling this is not an issue.

Our pulsed system has the capability of generating pulses between 70 and 740 ps. The basic laser system was introduced in [4]. We showed that the generated pulses are nearly Fourier transform limited [5]. The entire system consists of an ECDL with several fiber amplifiers in series. After the first fiber amplifier, the pulses are generated with an electro-optic modulator from the cw radiation. In the consecutive stages the pulses are amplified.

We focussed on increasing the output energy of the pulses while maintaining the spectral properties. To this end we have increased the efficiency of the first pulsed stage and added a second. Currently, our pulses have an energy of up to 2.6 \( \mu J \) depending on the pulse duration. In a first test of the second harmonic generation of the pulses we obtained conversion efficiencies exceeding 20 % (cf. fig. 1). Outgoing from that first test we anticipate that a third pulsed amplifier stage is necessary in order to fulfill the requirements.

![Figure 1: Second harmonic pulse energies (diamonds) and conversion efficiencies (circles) versus fundamental pulse energies.](image)

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S-EBIT facility: status report*

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S-EBIT [1] facility currently installed at GSI shall facilitate research and development works for SPARC experiments at FAIR. This accelerator-independent source of HCI shall provide ions necessary for R&D of HITRAP [2] experimental stations and serve as a standalone device for research and R&D activities (e.g. development of x-ray spectrometers, calorimeter detectors, x-ray optics etc. [3]). Furthermore, the combination of S-EBIT with the available laser infrastructure e.g. JETI200 will be a unique platform for the study of highly charged ions subject to intense laser radiation [4]. During the FAIR construction related shutdown period of the GSI accelerator complex, when little to no beam time can be provided, such an accelerator-independent source of ions is of particular importance (Fig. 1).

![Schematic of interrelated experimental arrangements](image)

Figure 1: Schematic of interrelated experimental arrangements (for description compare text).

The EBIT-I of the S-EBIT facility [5] has been successfully commissioned at the Helmholtz Institute Jena/GSI and can be stably operated. This EBIT is equipped with a few windows providing line of sight to the middle drift tube (trap) region at an angle of 90° with respect to the electron beam axis. On one of the windows a gas injection is realized in a colimated differentially pumped drift region, which is providing an atomic beam that is pointed directly on the electron beam. A buffer volume of the gas injection can be filled by a leak valve and/or a pulsed valve allowing for mixing a cooling gas. The trap pressure during the continuous gas injection is lower than 10⁻¹⁰ mbar. Another port, equipped with a Be-window, was used in order to characterize the EBIT by recording spectra of the x-rays emitted from interactions between the ions and electrons inside the trap region. A Si-pin diode detector (Amptek XR-100CR) with a measured resolution of about 300 eV at 10 keV was used for these measurements.

A typical x-ray spectrum with Xe and Ar injection is shown in figure 2 with the relevant peaks labeled. The spectrum was taken with the electron beam of a 10kV energy (and a current of 30 mA), which was compressed by 1 Tesla magnetic field provided by the superconducting coils. The Ar-K and Xe-L peaks can be seen along with the peaks due to radiative recombination (RR) into the xenon L-, M- etc shells.

![X-ray spectrum](image)

Figure 2: X-ray spectrum of Xe and Ar from the EBIT-I (for description compare text).

Currently a fast multi-parameter data acquisition system is being prepared for the future x-ray measurements that will combine the EBIT with a dedicated Si(Li) detector as well as with novel techniques based on combination of crystal- and microcalorimeter-spectroscopy up to the hard x-ray regime [3]. Moreover, a time-resolved x-ray measurements is being prepared, which is necessary for a characterization of the charge breeding process. This along with the time of flight (and/or magnet scan) for the evaluation of the charge state distribution of extracted ions is also vital for the experiments where the EBIT can serve as a source of ions, e.g. HITRAP, CRYRING.

References


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Ionoacoustic monitoring of high energetic ions at SIS-18∗

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As heavy ions are stopped in water, their energy is distributed in the form of the well-know Bragg curve. For a short pulse width, this can be considered a spatially confined, adiabatic heating, and the resulting temperature gradient results in a pressure wave containing temporal and spatial information about the beam puls. It has been shown before, that this signal is the spatial derivative of the energy distribution. Usually studied in the context of medical physics as an in-vivo range determination[1-3], in this work the feasibility of an ionoacoustic detector for high energetic ions has been investigated with 238Ur, 124Xe, and 12C at various energies and intensities.

A lead-zirconate-titanate crystal (PZT) ultrasound transducer (Videoscan, Olympus) was placed in a water basin axial to the beam facing a kapton entrance window. The transducer was positioned by a 3-dimensional motorized stage. After an amplification of 60 dB, the signals were acquired by a digital oscilloscope. With this rather high amplification, we were able to measure beam intensities down to 200 uranium ions, 5000 xenon ions, and 3×10^7 carbon ions per single pulse. The data acquisition was triggered by an accelerator “beam-on” signal (SIS18 extraction kicker).

A typical waveform is shown in figure 1. The signal can be separated in 3 distinct parts deriving from different source locations. A direct signal, which is the pressure directly from the distal Bragg peak front. Also, a reflection signal being the reflection of the pre-distal part of the Bragg peak at the entrance window. Between those two is a window signal, which is resulting from the temperature gradient at the interface between water and the entrance window. With this identification of the signal, the time differences between those signals multiplied by the speed of sound in water (1.488 mm/μs at 22°C) is the range in water. For a correct value of the speed of sound and density of the water, its temperature was measured during every shot. As a comparison value, we calculated the expected range of the used ions in water from a GEANT4 simulation including all used parts in the beam path.

As an example, selected results of all used ion types at 300 MeV/u for uranium and xenon, and 180 MeV/u for carbon are displayed in table 1. The measured data is very stable in time as indicated by the low standard variation σ, providing a sub-millimeter-precise measurement of the range over several shots. For all used energies and ion types, we are in good agreement with the simulated ranges.

The evaluation is ongoing and sophisticated methods to exploit the rich frequency content of the signals are being developed. With this further understanding, an ionoacoustic detector can be a simple, fast, and precise monitor for high energy ions even at high beam intensity, since this method is not suffering from e.g. EMP or pile-up effects as seen in standard particle detectors.

Table 1: Results from ionoacoustic range measurements.

<table>
<thead>
<tr>
<th>Ion type</th>
<th>Simulated range in mm</th>
<th>Ionoacoustic range in mm</th>
<th>σ in μm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium</td>
<td>14.28</td>
<td>14.43</td>
<td>74.5</td>
</tr>
<tr>
<td>Xenon</td>
<td>21.00</td>
<td>21.12</td>
<td>8.4</td>
</tr>
<tr>
<td>Carbon</td>
<td>70.43</td>
<td>71.03</td>
<td>7.7</td>
</tr>
</tbody>
</table>

Figure 1: Example waveform measured at a uranium beam with 300 MeV/u and about 200 particles per pulse. The distinct parts of the signal are indicated (see text).

References

Radioresistance of nucleobases to cosmic rays


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Introduction and Experiment

Complex organic molecules (COMs) have been observed in space, e.g., in interstellar clouds or Solar system objects such as comets. They can be synthesized following irradiation of ice layers on grains at low temperature by cosmic rays or Solar wind [1]. Organic matter is constantly delivered to earth from space (micrometeorites) and thus may have contributed to the emergence of life. It is thus of great interest to study the radiation resistance of COMs in order to determine their survival times in space.

In this project, we studied the radiolysis of nucleobases (adenine, cytosine, thymine, guanine) in solid phase at low temperature exposed to swift heavy ion beams. This allows us to simulate in the laboratory cosmic ray irradiation of COMs in space. The destruction of the initial molecules and the appearance of radiolytic products were monitored by in situ infrared absorption spectroscopy. Experiments were performed with 190-MeV Ca beams at the M-Branch of UNILAC as described in [2,3].

Radioresistance of Adenine

As shown in [1-3], the apparent disappearance cross sections $\sigma_d$ can be determined from the fluence dependence of molecular column densities. An important information, needed e.g. for estimating life times of molecules exposed to cosmic rays in space, is the dependence of the cross sections on the amount of deposited energy, as shown in Figure 1 for adenine. The cross sections are found to follow a power law as a function of the electronic stopping power, $\sigma_d \sim S_n^n$, with $n \approx 1.17$.

With this information, taking into account the flux distribution of cosmic rays, the half life time of molecules can be estimated [1, 3]. Deep inside a dense molecular cloud, where primary UV radiation cannot penetrate and only secondary UV photons induced by cosmic rays are present, the survival time of adenine exposed to cosmic rays would be of the order of 10 million years [3].

![Figure 1: Cross section for disappearance of adenine as a function of electronic energy loss.](image)

10 Mys is comparable to the lifetime of such molecular clouds. Therefore, it seems likely that adenine could survive in those "nurseries" of star formation. However, in the outer regions of dense clouds, photolysis by UV radiation is dominant. A further interesting finding is that a water layer on top of the adenine sample does not significantly reduce or enhance the radiosensitivity [3].

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Role of composition, bond covalency, and short-range order in the disordering of stannate pyrochlores by swift heavy ion irradiation*

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AXB2O7 materials with the pyrochlore structure exhibit complex order-disorder transformations in response to irradiation with swift heavy ions. Either an amorphous phase, a defect-rich fluorite-structured phase, or a mixture of both form within individual ion tracks. The propensity of a material to amorphize or to adopt the fluorite structure is typically attributed to the ratio of its cation ionic radii, rA/rB. To date, most work has focused on the zirconate and titanate pyrochlores (B = Zr, Ti), due to their widely differing ionic radii. However, both elements exhibit similar ionic bonding. Thus, the role of bond covalency on the radiation responses of pyrochlore materials remains unclear.

To elucidate this role of B-O bond covalency, various stannate pyrochlores (A = Y, Nd, Sm, Gd, Er, Yb; B = Sn) were irradiated with 2.2 GeV Au ions, as the Sn-O bond is highly covalent. The resulting structure changes were characterized by transmission electron microscopy (TEM), x-ray diffraction (XRD), and Raman spectroscopy [1].

Atomic structure in the ion tracks

TEM images showed continuous cylindrical tracks for all compounds. XRD revealed the loss of the pyrochlore structure with increasing ion fluence and the accumulation of amorphous or fluorite material, indicated by attenuation of the initial diffraction peaks and the growth of new peaks corresponding to the latter two phases.

The rate at which these new phases accumulate, as a function of ion fluence, is proportional to the fraction of each phase produced within an ion track. Because the cylindrical tracks are nearly homogeneous in the axial direction, these phase fractions can be expressed in terms of the cross-sectional areas of a single track that are transformed to each phase. Assuming that track formation occurs by amorphization following ion impact and subsequent recrystallization of the fluorite phase at the amorphous/crystalline interface, the following expression was developed:

\[ f_A(\Phi) = \frac{1 - e^{-\sigma_D a_\Phi + \sigma_\Phi_a}}{1 - \left( \frac{\sigma_D}{\sigma_A} \right) e^{-\sigma_D a_\Phi + \sigma_\Phi a}} \]  

where \( f_A \) is the amorphous phase fraction, \( \Phi \) is the ion fluence, and \( \sigma_D \) and \( \sigma_A \) are the cross-sectional areas for the defect-rich fluorite and amorphous phases, respectively. Fits of equation 1 to the phase fractions obtained from the XRD data are illustrated in Figure 1.

A clear dependence of the extent of amorphization on \( r_A/r_B \) is observed. The rate of amorphization in the stan-

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Figure 1: Cross-sectional areas of the amorphous and defect-rich fluorite regions as a function of \( r_A/r_B \).

Local ordering in the irradiated region

The initial Raman spectra of these materials correspond to pyrochlore structures, but they are modified by irradiation. At high ion fluence the spectra of all compounds become nearly identical(Figure 2), although the XRD results indicate different long-range structures. These results agree with recent study of the local structures of irradiated pyrochlores, which indicated short-range weberite-type ordering in those disordered to the fluorite long-range structure [3]. The spectra shown here are consistent with weberite-type local ordering. These results show that this local order is present not only in pyrochlores disordered to the fluorite structure, but also those that have been fully amorphized.

Figure 2: Raman spectra of irradiated pyrochlores.

Swift-heavy ion irradiation and annealing of A$_2$TiO$_5$ (A = Nd, Gd, and Yb)

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The structural responses of A$_2$BO$_5$ (A = Nd, Gd, and Yb; B = Ti) compositions irradiated with 2.2-GeV Au ions were investigated using transmission electron microscopy (TEM), synchrotron X-ray diffraction (XRD) and Raman spectroscopy. The extent of irradiation-induced amorphization of three titanate compositions is shown to depend on the size of the A-site cation, with smaller lanthanides leading to a lower susceptibility to the accumulation of radiation damage (Fig. 1) [1].

![Fig. 1: Calculated amorphous phase fractions as a function of fluence for three compositions. The lines represent the fit of the data to a direct impact model (Eq. 1: $f_A(\Phi) = (1 - e^{-\sigma \Phi})$). Error bars were derived from repeated deconvolutions of the XRD patterns.](image-url)

In the track-overlapping regime, complete amorphization occurs in all three compounds, despite the ability of Yb$_2$TiO$_5$ to incorporate a great deal of structural disorder into its initial defect-fluorite structure ($Fm\overline{3}m$). The reduced stability of Yb$_5$Ti$_2$O$_7$ (as compared with A$_2$B$_2$O$_7$-type compounds with similar structure) is attributed to the high cation stoichiometric ratio (A:B = 2:1), which results in a local distortion due to random arrangements of the dissimilarly-sized cations, as well as different defect energetics from the difficulty in fitting a large Yb in a small Ti site. Such stuffing of A-site cations causes the material to exhibit a defect-fluorite periodicity on a long-range, but a pyrochlore-like local ordering, which inherently leads to a lowered susceptibility to radiation damage. The morphology of the ion track, as observed from TEM indicates a disordered crystalline phase without a distinct core-shell structure (Fig. 2).

![Fig. 2: HRTEM image of cross section of single ion track revealing the heterogeneous damage morphology of Yb$_5$Ti$_2$O$_7$. The insets represent the FFT analysis of the in-track and surrounding matrix phases.](image-url)

Fully-amorphized samples were then isochronally heated at temperature intervals from 100 to 850°C. XRD analysis indicated a consistent damage recovery in Nd$_2$TiO$_5$ and Gd$_2$TiO$_5$, where both compositions recover their original structures ($Pnma$), with the degree of recrystallization being more efficient for the material that was more easily amorphized under ion irradiation. In contrast to the orthorhombic A$_2$TiO$_5$, Yb$_5$Ti$_2$O$_7$ exhibited recrystallization of a metastable, non-equilibrium orthorhombic phase prior to a transformation to the stable defect-fluorite phase. The multi-phase transition pathway can be attributed to the fact that the transition from the amorphous to the orthorhombic phase requires a lower activation energy than would the immediate formation of the stable defect-fluorite phase, due to fewer topological constraints from the smaller degree of atomic connectivity of the orthorhombic structure.


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Thermal defect annealing of swift heavy ion irradiated ThO$_2$*  

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ThO$_2$ has been proposed as a nuclear fuel alternative to UO$_2$ in which $^{232}$Th breeds fissile $^{233}$U. Thorium-based fuels offer several advantages over conventional uranium-based fuels, such as high chemical durability and improved thermophysical properties. Despite the international interest in ThO$_2$-based fuels, there exist few data regarding the structural stability of ThO$_2$ under extreme conditions encountered in a reactor core environment. In this study, the effects of dense electronic excitation from 2-GeV Au ion irradiation were investigated in order to simulate the electronic energy loss component from fission-fragment irradiation and elucidate the high-temperature stability of irradiation-induced defects [1].  

A compacted powder sample of microcrystalline ThO$_2$ was irradiated to a fluence of $1 \times 10^{13}$ ions/cm$^2$ with 2-GeV Au ions at the X0 beamline of the UNILAC at GSI. ThO$_2$ is non-amorphizable and does not undergo any phase transformations under swift heavy ion irradiation; therefore, local probe techniques were applied to characterize ion-induced structural disorder caused by point defects. The ion-irradiated sample was characterized using neutron total scattering and Raman spectroscopy as a function of isochronal annealing temperature.  

![Figure 1: Total scattering function of irradiated ThO$_2$ after annealing at different temperatures](image)

Neutron total scattering yields information from both Bragg and diffuse scattering. The Bragg contribution was characterized by Rietveld analysis of neutron diffraction and gave information about the crystalline fraction of the material. The diffuse signal represents the atomic disorder in the system. In order to characterize the disorder and defects that were produced in the irradiated sample, the level of diffuse scattering was monitored. Figure 1 shows the level of diffuse scattering, as indicated by the arrows, after heating the irradiated sample to different temperatures for 20 minutes. The diffuse scattering and shift in peaks in the diffraction patterns (not shown) demonstrated that a significant annealing event occurred after heating to 425 °C. Using Raman spectroscopy, which is sensitive to the CeO$_6$ polyhedra coordination environment, it was shown that the abrupt drop in diffuse scattering level is concomitant with a shift in the T$_{2g}$ Raman mode (see Figure 2 inset). Therefore, the structural change occurring between $\sim$300–400 °C is attributed to the annealing of small oxygen-interstitial-type defects.  

![Figure 1: Raman spectra of irradiated ThO$_2$ after annealing at different temperatures](image)

The defect annealing trend observed for swift heavy ion irradiated ThO$_2$ is in agreement with the defect annealing behavior of alpha- and neutron-irradiated UO$_2$ and PuO$_2$ [2]. In addition, the study demonstrated the ability to use neutron total scattering to investigate point defect phenomena in homogeneously-irradiated powders.

**References**  


Thermal diffusivity degradation of heavy ions irradiated isotropic graphite*

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Polycrystalline graphite (PG) is one of the most used materials for application in extreme radiation environments in high power accelerators because of its high thermal conductivity, thermal shock resistance and low activation. PG is considered as the material of choice for the beam catchers and the production target at the Super-FRS at FAIR. However, since radiation damage can cause severe structural changes, it is important to quantify the degradation of graphite properties under high intensity ion beam exposure.

Thermal diffusivity $\alpha$ is a material-specific property describing the rate of transfer of heat from the hot to the cold side of a material. Together with the specific heat $C_P$ and density $\rho$ it defines the thermal conductivity $\lambda$ by

$$\lambda(T) = \alpha(T) \times C_P(T) \times \rho(T).$$

The method for measuring thermal diffusivity was originally developed by Parker [1]. The rear side of a plane-parallel sample is exposed to a short laser pulse and the temperature rise on the front side of the sample is monitored by an IR detector. For thin and for high conductive samples the in plane measurement provides more reliable results [2]. A series of 20-mm PG discs with average thickness of 50 $\mu$m were irradiated at the M-branch of the UNILAC with 4.8 and 5.9 MeV/u Ca and Au ions. The average flux during irradiation was $2 \times 10^4$ ions/(cm$^2$s) and the accumulated fluence on series of samples was between $1 \times 10^{11}$ and $5 \times 10^{13}$ ions/cm$^2$.

The irradiation with Au ions leads to a significant drop of the thermal diffusivity, while lighter Ca ions have a smaller contribution at similar fluences.

Beam-induced structural changes of the samples were analysed by Raman spectroscopy (Fig. 2). The G-band is caused by vibrations of the ideal crystal lattice of graphite, and the D-band is due to defects in the material. With increasing irradiation the intensity $I_D$ of the D-peak grows. A quantitative analysis of the defect density $n_d$ is obtained by using the following formula [3], with $I_D$ being the G-band intensity:

$$n_d (\text{cm}^{-2}) = \frac{(1.8 \pm 0.5) \times 10^{22}}{\lambda_{\text{Laser}}^4} \left( \frac{I_D}{I_G} \right)$$

The increase of the defect density (Fig. 3) confirms the stronger effect of the Au+ ion irradiation on the degradation of the thermal diffusivity of polycrystalline graphite.

Figure 1: Thermal diffusivity of polycrystalline graphite as a function of fluence for Au and Ca ions. The dash-dotted line is an exponential fitting to the data.

Before and after irradiation the in plane thermal diffusivity of the samples was measured using a NETZSCH LFA42. Figure 1 shows the thermal diffusivity of irradiated graphite samples as a function of accumulated fluence.

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Figure 2: Raman spectra of PG irradiated with Au ions at different fluences. The spectra are normalized to the intensity of the G-peak.

Figure 3: Defect density of Ca and Au ions irradiated PG deduced from Raman spectroscopy data.

Study of high-energy ion beam induced dimensional changes in high density isotropic graphite

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Polycrystalline graphite (PG) is the material of choice for the target wheel and as beam catchers at the Super-Fragment Separator and for the beam dumps of experimental caves at FAIR. In the context of increased beam intensities and high accumulated doses, these FAIR components will experience structural changes \cite{1} leading to swelling and to thermal and irradiation creep. From low-doses neutron irradiation of graphite it is known that Frenkel pair generation leads to dimensional increase perpendicular and decrease parallel to the basal planes. The growth in normal direction to the planes is accommodated by the existing pores in the material. As a result, at low doses, the neutron irradiated isotropic graphite experiences an initial shrinkage. After most of the pores are closed, the swelling normal to basal planes starts to dominate and the graphite sample expands again until it finally fails.

The mechanical creep before the irradiation is much smaller compared to the beam-induced strain. During the irradiation, despite the applied load, a two-step shrinking process is observed. In the first step the dimensional change proceeds with a larger initial rate up to \(\sim 2 \times 10^{-12}\) ions/cm\(^2\). Above this fluence, the contraction rate decreases. This might be associated with clustering of irradiation-induced vacancies once a critical density is reached, leading to a decrease of the contraction rate or to an increase of the creep strain. Sudden elongations of the samples (e.g. at \(\sim 7.5 \times 10^{12}\) ions/cm\(^2\)) are associated with beam failures, as shown in figure 3. When the beam stops, the sample cools down leading to a small contraction. Afterwards the sample expands due to mechanical creep.

![Figure 1: Scheme of online experimental set up for creep measurements at cave A at SIS 18 accelerator at GSI.](image1)

![Figure 2: Strain as a function of accumulated fluence for an isotropic graphite sample applied to a tensile load of 5 MPa and exposed to 230-MeV/u \(^{238}\)U ions.](image2)

![Figure 3: Blow-up of variation of sample length during a beam failure interval; the accumulated fluence reached 7.3\times10^{12} \text{ U ions/cm}^2 at an applied tensile load of 5 MPa.](image3)

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References


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Upgraded in situ Raman spectroscopy system at UNILAC beamline M3*

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Introduction
Raman spectroscopy is widely used in materials research as well as in geosciences. It has proved to be an excellent method to analyze structural changes of solids exposed to high-energy ions available at large accelerator facilities. In situ investigations have the great advantage that the modification induced by the ion beam can be followed on a given sample spot by stepwise increasing the fluence without removing the sample from the irradiation chamber. This is of particular interest for natural materials that are often inhomogeneous and/or anisotropic. By analyzing the same spot after each irradiation step, crystal orientation effects are excluded because the sample remains in a fixed position.

A first version of a confocal Raman spectrometer was integrated at the M3 beamline of the UNILAC and successfully used for in situ analysis during beamtimes in 2014 and 2015 [1]. In the meantime several improvements were installed and tested during irradiation experiments in 2016.

System Upgrade
The main improvement of the upgraded system is a high-precision 3-axis stage consisting of three piezo stages (Physik Instrumente Q521-330) with a precision of 30 nm. The new stage has the advantage that it is equipped with a multiple-sample holder. This allows the irradiation of a series of samples without venting the irradiation/spectroscopy chamber and thus saves valuable beamtime. Moreover, it is now possible to precisely position the location of a sample spot to be analyzed. Also tiny samples such as for example nanofibers can be studied. For geological samples this is of great advantage in case of microanalysis of zoned samples.

In the upgraded version, the laser of the system is now operated by remote control providing a better possibility to monitor beam-induced ion-luminescence of samples. This allows us to cross-check the exact position and shape of the beam on luminescent samples like calcite (CaCO3).

First irradiations with 4.8 MeV/u Au ions in combination with Raman spectroscopy on calcite (CaCO3), dolomite CaMg(CO3)2 nanofibers and graphene were demonstrating the very useful improvements of the upgraded system.

References

Figure 1: a) Raman-System inside the irradiation chamber at the M3-beamline of the UNILAC, consisting of a Horiba Jobin Yvon iHR 320 spectrometer with a Pelletier cooled camera and a 532 nm laser (90mW power); b) new 3-axis piezo stage with a holder for several samples.

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Chemical track etching of ion-irradiated siderite

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In geosciences, fission-track dating is one of the main methods of thermochronological analysis. Mineral samples are etched to convert tracks of fission fragments into surface etch pits that can be viewed and quantified under an optical microscope. Below a mineral specific temperature, tracks accumulate over time due to the mineral’s natural uranium content [1].

Siderite (FeCO₃) is an important mineral formed during diagenetic processes and hydrothermal activity. In order to test the viability of track etching methods, we used siderite crystals from Grube Pfannenberger Einigkeit, Siegerland and irradiated them with 5.9 MeV/u ¹⁹⁷Au ions to a nominal fluence of 1 × 10⁸ ions/cm². During irradiation the crystals were covered with a hexagonal mask to create etched and non-etched areas on the crystal surface. A thin polycarbonate foil simultaneously irradiated was etched to determine the exact fluence applied.

Track etching of the irradiated siderite samples was performed in a solution of sulfuric acid (10%). The temperature of the etchant was thermally stabilized to 50 (± 0.1) °C using a heated water bath. The etchant was stirred with a magnetic stir bar.

The etch pits were counted and their dimensions measured using a reflected light Olympus BX 50 microscope and Stream Enterprise software. The etch pits exhibited a roughly hexagonal shape and reached a diameter of 3-4 µm after 60 min of etching (Fig. 1).

The analysis of several microscope images yields an area density of 2.14 (± 0.17) × 10⁶ etch pits/cm² for siderite and 2.10 (± 0.11) × 10⁶ pores/cm² for the polymer foil. Within the experimental error, the amounts of etch pits matches the fluence applied.

In conclusion, sulfuric acid is a useful etchant for visualizing ion tracks in siderite created by swift heavy ions. The etching solution applied converts all tracks into countable etch pits. Experiments with fission tracks induced by covering siderite samples with zircon grains, which emit fission fragments when irradiated with thermal neutrons, are in progress.


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Figure 1: Etch pits on surface of irradiated siderite (60 min, 50°C, 10% H₂SO₄, 20x magn., reflected light).

Figure 2: Etch pits on surface of irradiated siderite (60 min, 50°C, 10% H₂SO₄, 20x magn., transmitted light).

Figure 3: Etch pits on surface of irradiated polymer foil (90 min, 50 °C, 6 mol/l NaOH, 50x magnification).
The role of osmotic effects in asymmetric track etching

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The ion track technology provides unique opportunities for developing tailored micro- and nanoporous membranes with fine-tuned transport properties. In the last decade, special attention was given to so-called conical pores. Their fabrication includes the irradiation of a polymer foil with high energy ions followed by asymmetric chemical track etching. Recently it was evidenced that under such conditions an intense osmotic flux develops and has a strong effect on the nanopore geometry [1]. Here we report on experiments aimed at better understanding the role of osmotic phenomena in asymmetric track-etched nanopores.

Polyethylene terephthalate (PET) biaxially oriented foils (12-μm-thick Hostaphan RNK) were irradiated with 11 MeV/u Au ions at the UNILAC (GSI, Darmstadt) and with 1.2 MeV/u Xe and Kr ions at the IC-100 cyclotron (FLNR, JINR, Dubna). All samples were additionally exposed to ultraviolet radiation to stabilize the track-to-bulk etch rate ratio. Chemical etching was performed in an electrolytic cell at room temperature. One compartment of the cell was filled with 9 M NaOH and the other compartment contained a stopping solution (either neutral 1 M KCl or acidic 2 M KCl + 2 M HCOOH, 50:50, v/v). The cell was equipped with a capillary (2 mm inner diameter) inserted into the compartment with the alkaline etchant. The etched area of the foil samples was 1 cm². The geometry of the obtained asymmetric pores was investigated using field emission electron microscopy (FESEM).

Because the electrolyte concentrations on the two sides of the asymmetrically etched foil are significantly different, a gradient of the chemical potential of water generates a flow from the stopping solution to the etchant. Figure 1 shows the etching solution build-up in the capillary as a function of the etching time. For Au ion tracks, a pronounced osmotic flow starts at 90-100 and 140-200 min for neutral and acidic stopping solutions, respectively. Tracks of lighter ions (Kr) are etched through slower, however the resulting pores show a similar osmotic flow. The influence of acidity of the stopping solutions on the track etch rate indicates that unetched tracks are less permeable for electrolyte at low pH [2].

Due to osmotically driven flow of water into the pore, the alkali concentration gradient in the pore is essentially non-linear. The concentration is almost constant along the wide part of the pore and dramatically decreases in the pore tip region. This effect results in the conservation of the tip radius at a level of few nanometers after the etching and stopping solutions meet each other (see Fig. 2).

To our knowledge, clear osmotic effects caused by inorganic salts in track-etched membranes have not been observed before.

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Figure 1: Build-up of the alkaline solution level H as a function of time for asymmetric etching of PET foils with 10⁹ cm⁻² Kr (bottom) or Au (top) ion tracks. Solid and dashed lines show the respective data for neutral and acidic stopping solutions. Results of several parallel experiments for each ion are shown.

Figure 2: Track-etched pore tip ~20 min (left) and ~70 min (right) after breakthrough. One-sided etching of Au ion tracks continued after osmotic flow had been detected.

References

Nanoconfinement effects on solid-state, gel electrolyte systems *

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Recent findings demonstrating remarkable cyclability of MnO2 mesowires in gelled LiClO4-PMMA (poly(methyl) methacrylate) [1] motivated our research to understand how nanoconfinement affects the gel electrolyte. Because MnO2 possesses tendril and nanovoid structures, nanopores can be used to explore the effects of nanoconfinement on the LiClO4-PMMA gel electrolyte. In this report, we demonstrate a nanoscale solid-state ionic diode using LiClO4-PMMA gel electrolyte. This was accomplished by infiltrating a polyethylene terephthalate (PET) conical nanopore with LiClO4-PMMA gel. [2] Other experiments in cylindrical PET pores served to confirm fundamental characteristics of the ion transport in the solid electrolyte.

Our nanopores were fabricated by the track-etching 12-μm PET films irradiated at the UNILAC with single Au ions (11.4 MeV/u). Cylindrical pores were subjected to symmetric etch conditions while conical pores were prepared by asymmetric etch conditions. After etching, pores were characterized by I-V curves in LiClO4-propylene carbonate and then drop-cast with the LiClO4-PMMA gel.

Figure 1. I-V curves for cylindrical pores with both Li+ doped and undoped PMMA gels.

Initial experiments in cylindrical pores with the gel electrolyte demonstrated many properties of ion transport observed at the bulk scale translated to sub-micron confinement. Previous measurements of conductivity and ion transfer number [3] were confirmed in cylindrical pores by comparing doped and undoped (Fig. 1) PMMA gels. We also showed that PMMA fully infiltrated nanopores using simple drop-cast.

Experiments in conical pores demonstrated classic ion current rectification behaviors (Fig. 2). Current-voltage curves and long pulse (20 s) measurements confirmed the effect was robust. We hypothesize the macroscopic drop

of LiClO4-PMMA on the surface of the membrane served as an ion reservoir, preventing ion depletion for the long pulse measurements. Furthermore, we were able to demonstrate switching of the diode for a 0.1Hz, 2V amplitude square wave [2].

Figure 2. I-V curves from a 100mV/s pulse scan and a 20s long pulse measurement

The standard model for ion current rectification requires ion selectivity which, in nanopores, commonly results from presence of surface charge in nanoconfinement, leading to voltage-dependent conductive states. In the case of a solid-state electrolyte, elucidating mechanisms is not as straightforward, as no charges are expected on the nanopore surface. However, ionic selectivity can be contributed to other factors i.e. Li+ complexing with the PMMA to generate positive volume charge [4] (in agreement with Fig. 2) and confinement dependent conductivity [5].

These results detail the first, to our knowledge, nanoscale solid-state ionic diode and extends the range of electrolytes capable of testing with PET nanopores. Solid-state ion transport phenomena in nanoconfinement showed similarity to liquid counterparts, despite possessing different mechanisms for achieving them.

References

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Conical ion-track etched nanopores in SiO$_2^*$

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The ion-track etching method can be used to produce nanopores in a variety of materials. The aim of this work is to develop a better understanding of the ion-track etching process in SiO$_2$ and its dependence upon the un-etched track structure. To achieve this goal, we performed advanced synchrotron-based small-angle x-ray scattering (SAXS) measurements.

Films of 2 µm thick SiO$_2$ were irradiated with 1.1-GeV Au ions (10$^9$ cm$^{-2}$) at the GSI UNILAC accelerator and alternatively with 185-MeV Au ions at the ANU ion accelerator facility. Samples were subsequently etched in concentrations of 10, 5, and 2.5% hydrofluoric acid (HF) at room temperature for times ranging from 60 to 30 min.

After etching, some samples were investigated by scanning electron microscopy (SEM) revealing parallel oriented, conical pores of uniform size and morphology (Fig. 1). Obtaining cross-section images is, however, time consuming and the samples have to be broken.

Figure 2(a) shows the SAXS image of the same SiO$_2$ membrane SEM-imaged in Fig. 1. The complex image contains details about the pore morphology. Figure 2(b) shows the calculated fit to the image attained using a semi-analytical model for the cone form factor. This fit allows us to extract the length, opening angle and base radius of the etch cones averaged over millions of cones. Subsequently, we calculated the axial as well as the radial track etching rates. Figure 3 displays the base radius as a function of etching time obtained in 10% (blue), 5% (green), and 2.5% (red) HF. From the slope, a respective radial etching rate of 29, 12 and 7 nm/min is deduced. Plotting the cone length as a function of etching time (not shown), we obtain corresponding axial etching rates of 146, 62, and 33 nm/min. As expected the etching rates vary roughly linearly with etchant concentration.

Fig. 1: Cross-section SEM image of conical nanopores in SiO$_2$ obtained after 8 min etching in 5% HF. The pores display a base of ~160 nm and a very small tip size.

In contrast to SEM, SAXS is a non-destructive technique and it enables leveraging statistics, since many uniform structures are measured at once. Here, we apply SAXS to extract information about the morphology of the nanopores including base and tip diameters as well as opening angle of the cones.

Fig. 2: (a) SAXS image measured at the SAXS/WAXS beamline of the Australian Synchrotron; (b) calculated fit.

Linear extrapolation of the radial etch rates for the lower concentrations of 5 and 2.5% suggest non-linear behaviour at the early stages of etching because they do not intercept the radius axis at zero. The early stages of the etching process are interesting because the channel opening within the first few nm is influenced by the damage structure of the track. Planned experiments combining short etching times and in-situ SAXS studies will allow us to improve existing etching models and achieve a higher control on tailoring particularly small nanopores for specific applications such as nanoscale filtration and biosensing.

References


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The iNAPO-project: etched swift heavy ion damage tracks in polymers as biomimetic ion conducting nanopores for electrochemical sensors

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In nature, ion conducting nanopores play a vital role for the function of living cells. They undergo gating processes where they open and close upon an external stimulus, such as the presence of a particular biomolecule, the ligand. When the gating process is observed and is quantitatively measured, one can derive data about the presence and the amount of the ligand. Hence, the nanopores can be utilized for specific sensing. However, biological nanopores are embedded in a biological cell membrane that is fragile and unstable with respect to storage and application. The LOEWE project iNAPO (ion conducting NanoPOre) aims at combining robust polymer-based nanopores with protein-based biological nanopores, thus combining the selectivity and sensitivity of the latter with the stability and processability of the first ones. In the final step of the project, the biologically modified polymeric nanopores will be incorporated into a Lab-on-Chip system, creating a new type of device for applications, e.g., in medical diagnostics and environmental analysis. The whole bottom-up process is schematically shown in Fig. 1.

Figure 1: The iNAPO project: Fabrication steps of a micro sensing device for ion conducting nanopores.

The first essential step in the process of fabricating synthetic nanopores is the formation of a straight well defined damage track in a polymer. A single heavy ion, e.g. gold, accelerated in the UNILAC to a kinetic energy of several MeV/u, is directed perpendicularly towards the polymer foil, mostly polycarbonate or polyethylene terephthalate, that has a typical thickness of 10 to 20 µm. The ion passes the target in a straight line and producing a cylindrical zone of a few nm in diameter. This damage track consists of broken chemical bonds and reduced density. Due to the enhanced chemical reactivity, the track can be converted into an open channel by chemical etching in a suitable etchant cylindrical as well as conical channels can be produced. Their size is adjusted by the etching time and can be tailored between few nanometers and micrometers.

In the next step, the nanopore walls are chemically modified, i.e. specific molecules are attached to the surface. These molecules are able to specifically react with counterpart molecules, in a key-lock type reaction. This reaction is accompanied by a reduction in pore diameter. The process is monitored electrochemically by mounting the polymer foil (containing in most cases one single nanopore) between two compartments of an electrochemical cell. The cell is filled with an aqueous electrically conductive salt solution. When a voltage is applied between two electrodes placed in each of the cell compartments, an ionic current flows through the nanopore. When the nanopore surface is electrically neutral, the flow is the same in both directions, irrespective of the polarity of the electrodes. If, however, the surface is charged, the asymmetric shape of a conical nanopore results in a potential distribution yielding rectification, i.e. the ion flow in one direction is larger than in the other direction. The nanopore thus becomes a nanofluidic diode. The resulting current/voltage curve shows a steep line for a positive applied voltage and a flat one under negative voltage, or vice versa, depending on the sign of charge of the nanopore surface.

The following example of a biomolecular nanopore sensor deals with the analysis of pyrophosphate (P2O74–), an anion that plays an important role in biochemistry. In elevated quantities, it may cause various diseases, such as arthritis. Therefore, monitoring is of importance. For a nanopore-based pyrophosphate sensor, di(2-picolyl)amine groups are chemically coupled to the nanopore wall. Zinc ions are added and form a bis(Zn2+–DPA) complex. This complex specifically reacts with pyrophosphate while there is no reaction with other phosphates such as monohydrogen phosphate (HPO42–), dihydrogen phosphate (H2PO4–), and adenosine mono-, di- and tri-phosphate (AMP, ADP, ATP). Fig. 2 shows schematically the complexation reaction between the zinc-DPA complex and the pyrophosphate (shown in green).

Figure 2: bis[Zn2+–DPA] complexes (red) coupled to the nanopore surface selectively react with P2O74– (green).
The current/voltage curves in Fig. 3 show that the ion current through the nanopore is hardly changed when various phosphate ions are added to the electrolyte solution. In contrast, the addition of pyrophosphate causes a strong reduction of the current at negative voltages, and an increase at the positive voltage branch. This effect is due to the coupling of pyrophosphate with its 4 negative charges that influence the cation flow, leading to a diode-like effect. While the presence of other phosphates does merely change the nanopore current, pyrophosphate can be measured specifically, even down to submicromolar quantities. This example illustrates that ion-track based nanopores in polymer foils can be used as molecular sensors with a certain sensitivity and selectivity.

Figure 3: The ion current through the nanopore is changed in the presence of pyrophosphate (PP$_4$) but not when other phosphates (e.g., mono- and di-hydrogen phosphate or adenosine mono-, di- and tri-phosphate are added [1].

As mentioned before, an ongoing further step in the development is the combination of polymer nanopores with biological nanochannels. Figure 4 shows how binding proteins are used as a base for biological protein-based nanopores. In these hybrid pores, the second generation of functional nanopores, the distinct selectivity of the biological nanopores to ligands, i.e., the biomolecules to be analyzed, will lead to a further improvement in the functionality of the sensors.

So far, laboratory set-ups have been used for the electrochemical measurements. They consist of an electrochemical cell of some 10 to 15 cm length, connected via cables to a voltage source and a sensitive current meter. The project iNAPO comprises the fabrication of a compact lab-on-chip micro device with the polymer foil embedded in it by means of micro-nano-integration techniques. The polymer foil containing the nanopore is embedded into a microfluidic system with integrated electrodes. The foil is glued between two 200 µm thick sheets of an epoxy-based photoresist containing three microfluidic channels formed by photolithography. The lower polymer plate is placed on a glass substrate with two evaporated gold electrodes; the upper plate is covered with a glass plate. For operation, the microdevice will be connected to a power supply and an amperometer that are located in a compact unit where the microdevice is fitted in.

Figure 4: Schematic cross-section of a polymer nanopore with binding proteins included (dark blue) that are coupled to biological nanopores on top. These are gated by the presence of a specific analyte biomolecule (the ligand, reacting with the receptor) leading to an ion flux (red) [1].

Figure 5: Setup of microfluidic lab-on-chip system with integrated nanopore [1].

The microchannels are filled with electrolyte, and an amperometric reference value is recorded at a given voltage. Then, the electrolyte with the analyte molecules is added and the current response is measured again. In a calibrated system, the concentration of the analyte can be determined quantitatively, if it is within the measurement range of the system.

With such a compact set-up with low power input, it will be possible to carry out fast low-cost analyses, e.g., for medical diagnostics or for environmental tests.

Redox-responsive ionic conduction through single conical nanopores∗

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Ion channels are pores embedded in cell membrane that regulate the passage of ions and signaling molecules.[1] Natural ion channels are precisely controlled structures with defined interfacial chemistry. However, the fragility of the supporting lipid bilayer restrains their suitability for practical purposes. Conversely, synthetic nanopores have recently attracted interest because their geometry and surface pore properties can be tuned on demand.[2] Particularly, track-etched single conical nanopores (produced by irradiating polymer membranes with one individual ion at the UNILAC) exhibit remarkable transport properties such as permeability, voltage-dependent gating mechanism and current rectification, similarly to biological ion channels.[3] To date, a variety of responsive molecules and functional groups that respond to different external stimuli have been immobilized on the pore surface to tune ion transport properties. To broaden the scope of nanoporous systems, the synthesis and anchoring of more complex functional molecules, e.g., oxidation-reduction (redox) sensitive moieties, on the pore surface is a challenge for current techniques.

Here, we demonstrate the redox-sensitive ionic conduction through a nanofluidic diode based on single asymmetric nanopores fabricated in polyethylene terephthalate (PET) membranes.[4] To achieve this goal, 1-(4-aminobutyl)-3-carbamoylpyridinium (Nic-BuNH2) is synthesized and anchored onto the pore surface via carbodiimide coupling chemistry (Figure 1).[4] The single conical pore exhibits current rectification because of the ionized carboxylic group units on the pore surface (Figure 1a). The modification process resulted in the switching of pore surface charge from negative to positive due to the presence of positively charged quaternary pyridinium units (Figure 1b). This fact caused a reverse in the current rectification, i.e., the Nic-modified pore preferentially transported anions from the tip to the base (Figure 2A).[4]

Once the nanopore surface was successfully functionalized with nicotinamide moieties, we proceeded to study the redox reactions inside the confined environment. The immobilized nicotinamide moieties were in oxidized state (quaternary pyridinium form) and the net charge on pore surface was positive. For the reduction of nicotinamide moieties, the modified pore was exposed to sodium dithionite (Na2S2O4) prepared in sodium bicarbonate solution in dark for a time period. During the course of the reduction process, the oxidized (positively charged) pyridinium ring of nicotinamide was converted to 1,4-dihydropyridine (uncharged/neutral) form (Figure 1c). This process resulted in the loss of pore surface charges, and the nanopore behaved like an ohmic resistor as shown in Figure 2B. Hence, the reduction process switched the nanopore from an “ON” state characterized by a high rectified ion flux to an “OFF” state with a low non-rectified current. The dihydronicotinamide can be reoxidized to nicotinamide by exposing the modified pore to a solution of an oxidizing agent such as hydrogen peroxide (H2O2). The oxidation process resulted in the generation of positive charges on the pore walls, and concomitant current rectification inversion.

Figure 1: Scheme representing the changes in pore surface chemistry (a) as-prepared pore, (b) modified with nicotinamide moieties in oxidized state, and (c) pore with dihydronicotinamide moieties in reduced form.

Figure 2: (A) I–V curves of single conical nanopore (d ~ 24 nm and D ~ 400 nm) prior to and after modification with nicotinamide. (B) I–V curves of the modified pore exhibiting the switching of oxidized (positively charged) nicotinamide moieties into reduced (uncharged) states.

In summary, we have demonstrated the fabrication of a redox-sensitive nanofluidic diode whose transport properties can be tuned via oxidation and reduction processes occurring inside the pore. The redox reactions allowed us to reversibly switch the nanopore inner environment from hydrophilic and conducting (“ON” state) to a hydrophobic and nonconducting (“OFF” state).

References

Ion transport through single conical nanochannels synthesized by ion-track technology and atomic layer deposition of $\text{Al}_2\text{O}_3$, TiO$_2$ and SiO$_2$ *

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Solid state nanochannels exhibit unique transport properties for electrolytes including ion selectivity, ion current rectification and responsive behaviour to external stimuli such as pH value, temperature, or ion concentration [1].

The ion flux through a nanochannel can be described by nanofluidic models. At the nanochannel surface a layer of immobile and mobile ions is formed. The type of ions in this so-called electrical double layer (EDL) depends on the surface charge, which in turn is influenced by the pH value of the employed electrolyte. For each material, there is a specific pH value, called point of zero charge (PZC), where no surface charges and thus no EDL are present [2].

![Setup employed for $I$-$V$ measurements](image)

Figure 1: Setup employed for $I$-$V$ measurements (a) and $I$-$V$ curves recorded across single conical channels coated with a 15 nm thin layer of $\text{Al}_2\text{O}_3$ (b), TiO$_2$ (c), and SiO$_2$ (d). Each channel was characterized at three different pH-values (legend shown applies for b-d). The insets in (c) show typical curves for preferred anion- (top left) and cation-flow (bottom right).

Single conical nanochannels were fabricated by ion-track technology. This includes the irradiation of a 30-μm thick polycarbonate (PC) foil with one individual high-energy ion (e.g. 2-GeV Au ions) followed by chemical track etching. The irradiated PC foil containing the single ion track is mounted into an electrochemical cell exposing one side of the tracked foil, e.g., to a 9 M NaOH solution and the other side to water [3]. For this project, etching was performed at 30 °C for 15 min converting the ion track into an open conical channel with a base diameter of about 3 μm and a tip size of ~100 nm. Afterwards, the polymer foils were coated by atomic layer deposition (ALD) with 15 nm $\text{Al}_2\text{O}_3$, TiO$_2$, or SiO$_2$. The employed ALD reaction parameters have been previously adjusted to achieve homogeneous and conformal inner coating along the full length of the nanochannels [4].

Figure 1 shows the electrolytical cell (a) and the $I$-$V$ characteristics (b-d), measured across the single nanochannels using 1 M KCl solution with pH values of 2 (green), 5 (purple), and 9 (red) together with the linear $I$-$V$ characteristics measured across uncoated nanochannels (blue) at pH 5 (PZC of PC). The ion currents of uncoated channels are higher due to the larger tip diameter. After ALD, the $\text{Al}_2\text{O}_3$-coated channel (Fig. 1b) exhibits a linear $I$-$V$ curve at pH 9, which is the PZC of $\text{Al}_2\text{O}_3$ [5] while at lower pH values, the channel surface charges positively resulting in a preferential anion flow from tip to base. The SiO$_2$-coated channel (PZC = pH 2) [5] exhibits a linear $I$-$V$ curve at pH 2 (Fig. 1d). Increasing the pH value of the electrolyte increases the negative surface charges and the $I$-$V$ curve reveals a preferential cation flow from tip to base. Finally, the TiO$_2$ (PZC = pH 5) [5] coated channel exhibits symmetric ion flow at pH 5 (Fig. 1c). The surface charge can be either tuned positively (pH 2) or negatively (pH 9), resulting in either preferred anion- or cation flow.

After the ion transport measurements, each polymer foil was dissolved and free-standing, stable single conical nanotubes were imaged by scanning electron microscopy (SEM) (Fig. 2). The images reveal a conformal ALD deposition and a smooth nanochannel surface. We observed that the ALD coating improved the long term stability of the nanochannels, supposedly by prohibiting “closure” due to random polymer chain rearrangements.

![SEM images of free-standing single conical nanotubes](image)

Figure 2: SEM images of free-standing single conical nanotubes obtained by selectively dissolving the track-etched PC membrane after ALD coating with $\text{Al}_2\text{O}_3$ (a) and TiO$_2$ (b).

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Graphene/Polymer Composite Membranes by ion-track technology*


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Membranes can be used in a large variety of different applications, e.g., as separating agent for nano- and ultrafiltration processes [1]. The requirements for high-performance membranes are manifold including crucial characteristics such as excellent mechanical stability, high flow rates, and narrow pore-size distribution. Graphene, a monolayer of carbon atoms, exhibits all of these criteria. Due to its infinitesimal thickness of 0.3 nm and chemical inertness it is a promising material for filter applications.

In this work, we apply swift heavy ions (SHI) as an efficient tool to nanostructure graphene, i.e., to introduce defects of adjustable size and shape [2,3]. Our concept of fabricating high performance membranes is depicted in Fig. 1. A single graphene layer is transferred onto a mechanically stable polyethylene terephthalate (PET) support foil (thickness ~13 µm) which allows handling the membrane without damaging the graphene (Fig. 1a). By applying SHI irradiation, nm-sized pores are directly formed in the graphene and chemically modified tracks are created in the supporting polymer foil (Fig. 1b). By exposing the whole composite to a suitable etchant such as NaOH, the tracks in the PET foil are converted into open channels (Fig. 1c), which will serve as supply pipes to guide the fluid towards the nanoporous graphene where the filtering process takes place.

Figure 1: Schematic of fabrication process to produce graphene/polymer composite membranes: (a) SHI irradiation of graphene on PET foil, (b) generation of nanopores in graphene and ion tracks in PET foil, (c) converting ion tracks into open nanopores in PET by chemical etching. The graphene pores remain unaltered.

Figure 2 shows a representative scanning electron microscopy (SEM) image of a track-etched graphene/PET composite membrane. The sample was irradiated with 1.1-GeV Au ions at the X0 beamline of the UNILAC, applying a fluence of 1×108 ions/cm2. Subsequent etching was performed in a 3 mol/L NaOH solution at 50 °C for 50 min. Under these conditions, the ion-tracks were converted into open channels of diameter 640 ± 40 nm. The absence of charging effects during the SEM studies demonstrates the presence of the conductive graphene on top of the isolating PET foil. The inset presents a scanning force microscopy (SFM) image of a single ion-induced nanopore in graphene. The pore size distribution of the nanopores in graphene is narrow with a mean diameter of 12± 0.8 nm.

Figure 2: SEM image of a track-etched graphene/PET composite membrane. Inset presents a SFM image of a single ion-induced nanopore in the suspended graphene.

References


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0

2 µm

15 nm

S Both authors have contributed equally
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Lithographic editing-system based on track-etched conical pores in glass *

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A new lithographic editing system with the ability to erase and rectify errors on a microscale with real-time optical feedback is demonstrated. The erasing probe is a conically shaped hydrogel (tip size ~500 nm) template-synthesized from track-etched conical pores in glass wafers (Fig. 1). Our “nano-sponge” hydrogel probe “erasers” patterns by hydrating and absorbing molecules into a porous hydrogel matrix via diffusion analogous to a wet sponge. The presence of an interfacial liquid water layer between the hydrogel tip and the substrate during erasing enables frictionless uninterrupted translation of the eraser on the substrate. The erasing capacity of the hydrogel is extremely large due to large free volume of the hydrogel matrix. The fast frictionless translocation and interfacial hydration results in a high erasing rate (~785 \( \mu \text{m}^2/\text{s} \)) which is two to three orders of magnitude higher than AFM-based erasing (~0.1 \( \mu \text{m}^2/\text{s} \)) experiments.

The Polymeric Lithography Editor (PLE) pens and erasers were fabricated by means of template-assisted synthesis. In a first step, 70-\( \mu \text{m} \)-thick borosilicate glass substrates were irradiated with 2.2-GeV Au ions at the UNILAC of GSI. The created ion tracks were converted into conical channels by chemical etching in hydrofluoric acid. In a second step, a polydimethylsiloxane (PDMS) and agarose replica of the glass pore was generated.

Figure 1: Scanning electron micrographs of (a) cross-section of a single track-etched pore in a glass substrate. (b) PDMS replica fabricated by using a conical glass pore as template (tip diameter 500±100 nm, base diameter 22±1 \( \mu \text{m} \), pore length 38±1 \( \mu \text{m} \)).

The high precision and accuracy of our PLE system is obtained by coupling piezoelectric actuators to an inverted optical microscope (Fig. 2). Subsequently after erasing the patterns using agarose erasers, a PDMS probe fabricated from the same conical track-etched template was used to precisely re-deposit molecules of interest at the erased spots. The system also provides a continuous optical feedback throughout the entire molecular editing process including writing, erasing, and rewriting. To demonstrate its potential in device fabrication, we used PLE to electrochemically erase a metallic copper thin-film, forming an interdigitated array of microelectrodes for the fabrication of a functional micro-photodetector device (Fig. 2). High throughput, dot and line erasing, writing with the conical “wet-nano sponge” and continuous optical feedback, makes PLE complementary to the existing catalogue of nano/micro lithographic and 3D printing techniques. This new PLE technique will potentially open up many new and exciting avenues in lithography which have been unexplored due do the inherent limitations in error rectification capabilities of the existing lithographic techniques.

Figure 2. (a) Schematic of custom built PLE system with XYZ stage coupled to an inverted optical microscope. (b) Linear increase of the pattern diameter (in \( \mu \text{m} \)) with the square root of the erasing time (in s). (c) Schematic of absorption of molecules upon contact with the PLE tip with a contact time of ~1.8 s. (d) Fluorescence micrograph of PLE erasing with a 5 w/w% of agarose after gelation. The diameter of the erased patterns is 11.7 ± 0.7 \( \mu \text{m} \). (e) Continuous line deposition with agarose PLE equilibrated in a 2% fluorescein solution. (f) Schematic of line pattern deposition of fluorescein using agarose encapsulated with fluorescein. Both the deposition and erasing occurred via the water meniscus formed upon contact between the conical hydrogel tip and the substrate. Scale bar in (e) and (d): 30 \( \mu \text{m} \).

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Seebeck coefficient measurements of Bi$_{1-x}$Sb$_x$ nanowire arrays

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Thermoelectric (TE) devices are considered attractive potential pollution-free sources of energy, since they are able to convert thermal energy into electrical energy by means of the Seebeck effect. Moreover, supplying current through a TE device generates a temperature gradient by means of the Peltier effect, making TE devices suitable also for applications e.g. in cooling systems. The efficiency of a TE material is described by the figure of merit $Z = S^2/\kappa$, where $S$ denotes Seebeck coefficient, $\sigma$ electrical and $\kappa$ thermal conductivity, the latter including contributions of charge carriers and phonons. Theoretical calculations predicted an enhancement of the TE efficiency in Bi$_{1-x}$Sb$_x$ nanowires (NWs) due to the presence of finite-and quantum-size effects compared to the bulk systems [1]. Moreover, a further enhancement of $ZT$ is predicted when decreasing the NW diameter and adjusting the composition. In fact, a high value of $ZT \approx 2.5$ was calculated for 40 nm diameter wires with 13% Sb concentration. Despite these promising predictions, comprehensive and systematic measurements of the TE properties of Bi$_{1-x}$Sb$_x$ NWs as a function of wire diameter and orientation are scarce. The difficulties include the synthesis of Bi$_{1-x}$Sb$_x$ NWs of controlled diameter and the task to contact single NWs to measure the value of $S$, $\sigma$ and $\kappa$ without affecting their chemical and mechanical properties [2]. Here, we present measurements of the Seebeck coefficient of arrays of Bi$_{1-x}$Sb$_x$ NWs with controlled composition and diameters ranging from ~750 down to ~40 nm.

Polycarbonate foils (Makrofol N, thickness 30 $\mu$m) were irradiated with ~2 GeV Au ions at the GSI UNILAC. Track etching in a 6M NaOH solution at 50°C, converted the tracks into open pores with controlled diameter between ~40 and ~750 nm. The Bi$_{1-x}$Sb$_x$ NWs were subsequently electrodeposited in the pores using a three-electrode cell with a saturated calomel reference electrode at room temperature. The electrolytes were based on hydrochloric acid and different concentrations of Bi(III)- and Sb(III)-chloride [2,3].

Since the TE properties of the synthesized Bi$_{1-x}$Sb$_x$ NWs depend on both crystallographic orientation and composition, systematic investigations by both X-ray diffraction (XRD) and energy dispersive X-ray analysis (SEM-EDX) were performed as a function of electrolyte composition (i.e. using different concentrations of Bi(III)- and Sb(III)-chloride) and average wire diameter. The results revealed that both crystallographic orientation and composition do not vary significantly as a function of wire diameter. Subsequently, different arrays of Bi$_{1-x}$Sb$_x$ NWs were inserted in a setup specifically developed for measurements of the Seebeck coefficient from room temperature down to 20 K.

Figure 1 shows the Seebeck coefficient values measured on Bi, Bi$_{0.85}$Sb$_{0.15}$ and Sb NW arrays at room temperature as a function of the wire diameter. The data allow us to distinguish three different regimes: (Zone 1) wire diameter larger than ~130 nm, $S$ is similar to values of bulk materials (red area); (Zone 2) when decreasing NW diameter from ~130 nm to ~60 nm, $S$ decreases monotonically (green area); (Zone 3) for nanowires with diameter below ~60 nm, the absolute value of $S$ increases with decreasing wire diameter (blue area).

Given that composition and crystallographic orientation of the NWs do not vary significantly as a function of wire diameter, we attribute the monotonic decrease in zone 2 to the complementary contribution to the total Seebeck coefficient of the wire of both bulk- and surface states. The analysis of the data using a two-channel model supported this assumption [4]. Only for wires thinner than ~60 nm the Seebeck coefficient raises possibly due to quantum size-effects on both NWs surface and bulk states of the nanowires.

To conclude, these measurements demonstrate the possibility to tailor and eventually further increase the Seebeck coefficient in thin nanowires due to modifications in the electronic structure of both surface and bulk states of the material. This may lead to a further increase of $ZT$ for even thinner wires, making them suitable for thermoelectric applications such as sensing.

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267
A new setup for the investigation of stimulated desorption

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A new laboratory setup for investigations on beam and temperature desorption processes was established. To compare the amounts of released gas, different stimuli such as thermal desorption, ion-, electron- and laser-induced desorption are possible. During future beamtimes the setup is planned to be implemented into the M3 beamline at the UNILAC or the CRYRING.

During the 2016 beamtime with swift heavy ions it was found ion-induced desorption to originate not only from the surface, but diffusion from the bulk is also significant [1, 2]. One aim of the new setup is to investigate the contribution of the bulk more detailed. The investigations with different stimulations allow to distinguish the influence of surface and bulk separately, because of different ranges of ions and electrons. While running the setup on the accelerator, ion-induced desorption at different sample temperatures and beam parameters will be investigated. Additionally measurements on cryogenic surfaces are possible. By varying the thickness of frozen gas layers, the influence of the substrate to desorption can be analyzed.

The setup (figure 1) consists of two vacuum chambers, which are separated by an aperture. The temperature of this section must be constant to reduce thermal desorption form the aperture. Therefore it is water-cooled and a smaller aperture, made out of ceramics, is mounted to decrease the thermal conduction. For the measurements, the sample is moved in front of the aperture, so it is closed by the sample and only molecules desorped from the sample surface get into the measuring chamber. For the analysis a residual gas analyzer with m/q up to 200 and a sensitivity of 5 · 10⁻¹⁴ mbar ist used. The sample can be fixed on a heating assembly or a cryostat. Thus, a temperature range from 10 K up to 800 °C can be applied. In addition there are flanges for the connection to the accelerator and the installation of an electron and a keV-ion source. A pumping system evacuates the setup to a pressure of 1 · 10⁻¹⁰ mbar. A load lock for quick sample exchange is included.

First investigations on thermal desorption spectroscopy (TDS) are shown in figure 2. The gases desorb on different temperatures. Hydrogen has the highest desorption temperature due to diffusion from the bulk. In contrast the other gases desorb form layers close to the surface and show lower desorption temperatures [3].

The measurements on thermal desorption spectroscopy will be continued. In future the cryostat, the electron and the ion source will be mounted to do investigations on electron- and ion-induced desorption.

References


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Figure 1: Schematic sketch of the installed setup. The cooling / heating assembly denotes either the heater or the cryostat. The loadlock is not displayed.

Figure 2: First measurement of thermal desorption from a copper sample.
Thermal pre-treatment to minimize ion-induced desorption

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Heavy ion-induced desorption is a major intensity limitation for particle accelerators. Due to interaction of the particle beam with gas molecules, ions can hit the beam tube wall and stimulate the release of gas. At high beam intensities it is of most interest to develop new solutions to minimize the amount of desorbed gas molecules per incident ion, the so-called desorption yield.

In 2015, we tested the desorption yield (desorbed gas molecules per incident ion) for different accelerator-specific materials exposed to different cleaning procedures and revealed thermal annealing as the most effective treatment [1]. Last year we have systematically investigated desorption under thermal annealing between 200°C and 600°C for various annealing times [2]. The reduction of the desorption yield can be described as an exponential decay as:

\[ \eta = \eta_0 + a e^{-bX} \]  

with X being the annealing time or the annealing temperature, \( \eta_0 \) is the ultimate desorption yield value and a and b are constant values. The decrease of the desorption yield as a function of annealing time at 200°C is shown in figure 1. At this temperature the yield decreases down to about 18% of the initial value. The annealing affects all desorbed gas species, H₂, CO and CO₂ to similar amounts.

Increasing the annealing temperature also leads to an exponential decay of the desorption yield as shown in figure 2 for oxygen-free copper samples, all annealed for 24 h. Also here, a limit is reached at 550°C, where an increase of the temperature is not further effective. However, the desorption yield can be reduced down to about 3% of the initial value which means an important improvement for dynamic vacuum effects in particle accelerators.

Figure 1: Normalized desorption yield of gold-coated copper as a function of annealing time. The annealing temperature for each sample was 200°C. \( \eta_0 \) indicates the saturation value compared to the initial desorption yield.

Figure 2: Normalized desorption yield as function of annealing temperature. The annealing time was 24 h for each sample.

References


ESD setup for desorption yields investigation on various metals

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Introduction
A new ESD (electron stimulated desorption) experimental setup has been developed at the vacuum lab CSVS (Common System Vacuum System) Division. The existing setup from previous work [1] has been reconstructed, in order to investigate the desorption yields of various metals used in UHV (ultra-high vacuum) chambers. The desorption yield plays an important role in understanding the interactions between the chamber surface and charged particles such as ions and electrons.

ESD Experimental Setup
The new ESD setup consists of two vacuum chambers separated by a ceramic insulator. The first chamber consists of a pumping system with a known aperture, residual gas analyser, extractor gauge, and gas inlet. The second chamber is dedicated for loading and placement of the sample. Figure 1 shows a schematic of the configuration of the ESD experimental setup.

Figure 1: Schematic configuration of the ESD experimental setup at Vacuum Lab CSVS Group.

The main new feature of this setup is a new sample holder that has been designed in such a way to allow investigation of desorption yields of various metals more efficiently. Figure 2 shows the sample holder involving the filament, sample head, and electrical feedthrough.

The filament used for this work was a tungsten filament as electron source. The sample, located at the sample head, was placed above the filament and biased at positive voltage to attract the electrons from the filament. The electrical feedthrough on the sample holder allowed a current flow through the filament, voltage bias on the sample, current measurement on the sample, and ground connection.

The current through the filament was normally around 3 Amp, 9 Watt power. The voltage for the sample was biased from +0 V to + 500 V. The current due to the electron bombardment on the sample was measured using a Picoammeter.

Figure 2: A new design of sample holder for the ESD experimental setup at Vacuum Lab CSVS Group.

Figure 3 shows preliminary data obtained from the ESD measurement. As shown in Figure 3, the ion current measured on the samples was polarised into minus current due to the electron bombardment on the sample. At the same time, there was a change on the ion current for residual gas composition and the chamber pressure. The desorption yield can be calculated from the data of ion current and partial pressure measurements. This will be conducted in the future work.

Figure 3: Preliminary results showing the effect of electron bombardment on the sample, chamber pressure, and residual gas compositions.

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Operation and improvements of PHELIX

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The Petawatt High Energy Laser for heavy Ion Experiments (PHELIX), a user facility of GSI, has been serving an international user community for nine years as the main experimental capability of the plasma physics department. PHELIX is a versatile nanosecond and sub-picosecond laser delivering pulses above 100 J and in a peak-power range up to 500 TW. In 2016 a major upgrade of the pre-amplifier section has been performed without a significant impact of the beam-time: As planed, ten experiments were conducted. Three of experiments were made at the UNILAC experimental area, where one was done in combination with the ion beam available at this site. The rest was done at the in-house experimental area at the PHELIX building. Details about the various experimental beam times delivered by the laser are described in the plasma physics section of this annual report [1 ... 9]. Additionally, one internal experiment was done in-house. Basing on results of experiments performed at PHELIX, several peer-reviewed articles were published in 2016 [10 ... 16]. Since it takes time to publish results afterwards, most of these took place in the year(s) before. One of the worldwide unique aspects of PHELIX lies in its very good temporal contrast, whose characteristics and most recent developments have been published in [17].

Operation and improvements

As illustrated in Fig. 1, about half of time was spent to prepare and to perform external and internal beam-times. The PHELIX shot data base registered 414 shots on target. 5.1 % (in total: 21) of these are marked as failed: 16 due to problems with hardware, four caused by the experimentalists, one due to an operator failure. The experimentalists got compensation for all failed shots. Most of the shut down time was spend to rearrange the pre-amplifier section. Details can be found in [18]. Additionally, simulations were started to develop a new main amplifier head design for higher repetition rate operation [19], and two in-house developed measurement devices were brought to daily operation [20, 21].

Outlook

The increase of the repetition rate of the pre-amplifier section to somewhat of a few shots per minute for dedicated high repetition rate experiments with energies at the Joule level will be continued after the optical setup has been changed successfully.

Figure 1: Usage of PHELIX in 2016

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Implementation of the upgraded PHELIx pre-amplifier *

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In the past year, significant progress has been achieved at the PHELIx laser facility on the way towards higher pulse repetition rates. After detailed studies on the characterization of a flashlamp-based amplifier head and improvements of the optical layout the new pre-amplifier system has been implemented at the PHELIx facility. This step represents an important milestone on the way towards higher performance in terms of output energy and shot repetition rate.

The PHELIx Pre-Amplifier is a flashlamp-based Nd:glass laser amplifier system with a typical pulse output energy of up to 10 J and a repetition rate of one shot every 3 minutes. This limitation mainly arises from thermal wavefront deformations such as defocus, astigmatism, and higher order aberrations as well as birefringence effects. However, some experiments using only the pre-amplifier would greatly benefit from a higher repetition rate. In addition, the design of the 100 J laser to be built for the plasma physics collaboration at FAIR includes a pre-amplifier with the same characteristics as the one of PHELIx but this amplifier should operate at a higher repetition rate. For this reason, it was decided to upgrade the PHELIx pre-amplifier to a repetition rate of 3 shots per minute not only to serve the greater user community but also to gather hands-on experience for the FAIR 100 J laser.

The project follows two lines: at first, a new amplifier layout had to be implemented to overcome the thermal loading limitations of the previous design and second, individual components have been improved. The 45 mm amplifier has been redesigned, which includes a new 45 mm head as well as a new power supply using low voltage (2 kV) and off-the-shelf components. The newly designed optical layout consists of two amplifier heads (19 mm and 45 mm diameter) in double-pass configuration combined with Faraday rotators (Fig. 1 top). The new system needed to be inserted in an existing laser system which led to many layout constraints such as the position of object and image planes, input and exit beam diameters as well as various space limitations.

In a two month shutdown phase in summer 2016, the new system was successfully installed (Fig. 1 bottom). First measurements demonstrated output energies up to 17 J with considerable margin in terms of flashlamp voltages. Hence the design value of 20 J should be easily reachable. In addition, particular attention was paid to the optimization of the passive transmitted wavefront through the system. Precise alignment in combination with a strongly enhanced Shack-Hartman wavefront sensor (developed at PHELIx) allowed for an overall wavefront error of less than λ/4. After correction of astigmatic aberrations using controlled deformation of a polarizer plate in the beam path the residual wavefront error corresponds to spherical aberration as the most prominent contribution. During a shot the defocus aberrations is by far the dominating contribution to the wavefront deformation. This effect is compensated using a motorized lens at one of the relay telescopes. Higher order aberrations are compensated by a deformable mirror.

First tests also showed that the magnification of a telescope led to unexpected aperture clipping at the 45 mm head which lead to very little alignment margin and turned out to be critical for daily system usage. Hence, slight modifications of the magnification of telescopes before and after the 45 mm head will easily overcome the limitation. The new lenses will be installed in an upcoming maintenance period. Meanwhile several beamtimes have been performed since the implementation of the new system. The next step will focus on an operation at higher repetition rates on the order of one shot every 20 to 30 s.

Figure 1: The new PHELIx pre-amplifier as planned (top) and implemented (bottom).
Development of an actively cooled glass amplifier at PHELIX

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To improve the repetition rate of the High Energy Laser for heavy Ion Experiments (PHELIX) a cooling system for the main amplifier is required. In preparation for this task several simulations have been conducted to optimize the coolant flow, identify promising cooling agents and to improve the cooling time by using a time dependent temperature profile for the coolant.

Coolant flow

To ensure the wavefront of the Laser to be as undisturbed as possible after passing through the amplifier, it is important to reduce temperature gradients inside the glass to a minimum. This can only be achieved if the coolant is applied to the glass homogeneously. Using CADFEM ANSYS to run 3D fluid dynamics simulations it was possible to develop a concept to distribute the coolant evenly over the total width of the glass plate as can be seen in Figure 1. To facilitate the removal of potential bubbles the coolant was injected from the bottom and extracted from the top while the overall design is a sandwich of glass - coolant - glass.

![Figure 1: Fluid velocity inside the amplifier](image)

Coolant temperature profile

Simulations have shown that the thermal conductivity of glass is too low to achieve the goal of reducing thermal gradients below 0.01 K within 10 minutes if the coolant is injected at a constant temperature. To counter this problem we devised a plan to inject the coolant at a lower temperature during the initial phase of the cooling cycle to induce higher thermal gradients between the glass and the coolant and thus increase the thermal flux. To stabilize the temperature at the specified design point of the amplifier the coolant temperature is raised to the design point temperature in the second phase of the cooling cycle. This concept was again simulated using CADFEM ANSYS (an example can be seen in Figure 2) and the durations and temperatures of the phases were optimized.

![Figure 2: Temperature distribution inside the glass and fluid (cut view from the side @ t = 10s)](image)

Wavefront construction

Based on the previous simulations it is possible to calculate the deformations of the wavefront caused by the remaining thermal gradients left in the glass after one cooling cycle. This is caused by the differences in the optical path due to the thermal dependency of the refractive index and the thermal expansion of the glass. The constructed wavefronts were used as a benchmark for comparing different cooling agents and to further improve the temperature profile of the coolant.

Outlook

The development of the amplifier requires experimental data to complement the information gained from the simulations. Therefore experiments have been planned to test the chemical compatibility of the cooling agent with the glass as well as improving the cooling of our flash lamps and develop a design for the amplifier that is coolant tight.
Noise reduction technique for high dynamic range temporal laser pulse profile measurement

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At the PHELIX laser a novel technique is proposed which enlarges the dynamic range of temporal laser intensity pulse profile measurement devices by several orders of magnitudes.

In today’s laser-plasma experiments it is essential to know the whole temporal intensity profile of a high intensity laser pulse. The intensity range of interest spans over twelve orders of magnitude, from the ionization threshold of matter, around $10^9$ W/cm², up to the maximum achievable peak intensity. Unfortunately available measurement devices are limited to nine to eleven orders of magnitude dynamic range. Temporal high dynamic range measurements are typically done by third order cross-correlators, a scheme of such a device is shown in fig. 1. The limits for the dynamic range are formed by the maximum signal strength and noise photons, generated in the sum-frequency-generation (SFG) process.

![Figure 1: Scheme of a third order cross-correlator.](image)

Noise photons can be generated by interaction of the beam of the fundamental frequency $\omega$ with itself. This is a two step process, illustrated in fig. 2, where parts of this beam are frequency doubled and scattered into the direction of the frequency doubled beam. In this case the same optimized conditions apply for the SFG of such a frequency doubled, scattered photon with the fundamental beam to noise as it applies for the SFG of the signal.

Calculations show that the signal to noise ratio (SNR) depends on the phase mismatch of the frequency doubling ($\Delta k$), the nonlinear optical coefficient ($d_{eff}$) of the $1\omega$ beam in the recombinant crystal and the scatter distribution $S$ of the generated $2\omega$ noise light into the direction of the $2\omega$ beam, which all depend on the angles of incidence of the fundamental and frequency doubled beam $\alpha_1$ and $\alpha_2$, respectively, and the cutting angle of the crystal $\Theta$.

$$SNR = \frac{I_{1\omega}}{I_{2\omega,\text{noise}}} \propto \frac{\Delta k^2}{S d_{eff}}$$ (1)

![Figure 2: The left side of the scheme shows a nonlinear crystal and the both interacting beams. The right side shows a magnified area of the fundamental beam ($1\omega$), which generates frequency doubled photons ($2\omega$) and interacts with these photons to noise photons (noise).](image)

To maintain phasematching for the SFG of the signal these three angle have to be matched to each other. With this the increase of SNR depending on the angle of incidence $\alpha_1$ was calculated. The result is shown in figure 3.

A prototype device was built to validate this technique. The published results [1] show the first measurement of a laser pulse over 12.5 orders of magnitude and therefore experimentally validates the proposed technique.

References

Improvement of the homogeneity of the laser-driven proton beam within the LIGHT project

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Laser-driven ion acceleration is an emerging and promising field in which the Laser Ion Generation Handling and Transport (LIGHT) project makes a significant contribution. LIGHT combines the laser-driven ion acceleration with conventional accelerator technology realized in a worldwide unique test beamline [1] and leads to developments of experimental and applied science capability for e.g. FAIR and radiography. The project is based on a Target Normal Sheath Acceleration (TNSA) source driven by the Petawatt High-Energy Laser for Heavy Ion Experiments (PHELIX) 100 TW beam resulting in a continuous proton energy spectrum. The generated ions are captured by a high-field solenoid for energy selection of protons of \( 9 \pm 1 \) MeV via chromatic focusing and transported into a radiofrequency double spiral resonator operating at -90 degrees synchronous phase which is used for phase rotation of the single ion bunch. Behind the resonator, the beam is transported into a second target chamber. This transport can optionally be supported with two permanent quadrupole doublets. In the second target chamber a second high-field solenoid system was installed for steep focusing to access highest proton peak intensities. The beamtime in April 2016 aimed to improve the proton beam homogeneity. This goal is essential to enable the time-resolved imaging capability of the laser-driven proton beam and for the determination of a density distribution of a sample. In this beamtime, the so called Radiochromic Imaging Spectroscopy (RIS) was chosen as the detection method in which several radiochromic films (RCF) enable a spectral and spatial analysis of the beam profile. Due to the Bragg behaviour of the protons connected to a corresponding position in the film, an energy resolution is possible. Based on the transverse spectrum, the so called normalized beam uniformity factor (isnorm 13694:2015) can be calculated, which describes whether the beam has an uniform distribution (U_\eta = 0 completely uniform).

We have shown already the successful generation of sub-nanosecond focused proton bunches [2], this time compression enables the resolution of fast dynamic processes. Now we will improve the beam homogeneity. Figure 1 a) shows the beam profile recorded on a RCF film (Bragg peak at 7.4 MeV) at 6 m distance from target and shows low particle numbers but has a good homogeneity (U_\eta = 0.27). Based on TraceWin simulations, the removal of the quadrupole doublets within the beamline improved the transport efficiency. Figure 1 b) demonstrates a higher energy deposition at 6 m distance from target and a star shaped inhomogeneity leading to U_\eta = 0.50. In the next step, a 1.25 μm thin mylar foil for transverse scattering (negligible energy loss of 10%) was placed between the first solenoid and the rf cavity leading to significant improvements. Figure 1 c) shows the beam profile of a beam which is compressed in time with a mylar foil, d) energy compressed beam with a mylar foil.

Figure 1: Beam profiles at 6 m distance from the target: a) beam with the installed quadrupole doublets, b) beam without the quadrupole doublets, c) time compressed beam with a mylar foil, d) energy compressed beam with a mylar foil.

References
Further steps towards the generation of intense, subnanosecond heavy ion bunches at LIGHT *

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The LIGHT collaboration has been founded to provide a testbed for Laser Ion Generation, Handling and Transport [1]. The laser ion generation is based on the Target Normal Sheath Acceleration (TNSA) mechanism and is driven by the PHELIX 100 TW beam line at GSI. A pulsed solenoid captures and collimates a part of the divergent ion beam with a continuous energy spectrum by means of achromatic focusing. The resulting collimated beam can be compressed in phase or energy in a radiofrequency (rf) cavity, which is situated two meters behind the ion source. The resulting ion beam is then diagnosed with a diamond detector for a temporal depiction of the achieved phase focus at a distance of six metres from the target.

After a successful first demonstration of the generation, handling and transport of fluorine ions in 2015 [2] a subsequent campaign was launched in 2016 to optimise the resulting ion beam. One of the main differences to the generation of intense subnanosecond proton pulses [3] using the same setup is, that the central energy of the transported fluorine ion bunch is only a tenth of the central energy of the proton bunch. Therefore the fluorine ion beam is longitudinally much longer at the entrance of the rf cavity than the proton beam and exceeds the cycle duration of the rf cavity. This leads to the formation of multiple ion bunches with varying degrees of phase compression.

The utilised rf cavity is a three gap spiral resonator and is designed for particles with kinetic energies of 8 MeV/u. The effectiveness of the bunching depends on the kinetic energy of the particles which can be seen in figure 1. The gap voltage in gap two has a phase difference of \( \pi \) with respect to gap one and three. This means for maximum effectiveness, the particles must have travel times from gap to gap of \( 9.2 \cdot (n+1)/2 \) ns (standing waves \( f_{rf}=108.4 \) MHz).

To reduce the number of intense ion bunches one can exploit the characteristics of the cavity. By increasing the energy of the collimated F\textsuperscript{7+} ions from 0.95 MeV/u to 1.31 MeV/u and adjusting the rf power to achieve temporal focussing one strongly overcompensates the temporal divergence at lower energies and undercompensates at higher energies. The result of such a configuration can be seen in figure 2, where also the result of 2015 is depicted. It shows, that in the campaign 2016 we were able to suppress all other bunches when compared to the maximum intensity bunch at around 1.3 MeV/u. The steep slope of \( U_{eff} \) equals a high sensitivity of the minimum FWHM temporal width to the phase of the cavity and therefore the high intensity bunch is still in the order of nanoseconds.

![Figure 1: The energy necessary to achieve a temporal focus of F\textsuperscript{7+} ions is plotted as \( \Delta E \) (distance between the circles indicate \( \Delta t=9.2 \) ns at cavity entrance), whereas \( q/m \cdot U_{eff} \) demonstrates the effective gap voltage for F\textsuperscript{7+} ions.]

![Figure 2: Time of flight data of diamond detector converted to energy per nucleon]

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Energy selective focusing of TNSA proton beams by picosecond-laser driven ultra-fast EM fields∗ †

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Abstract

This work demonstrates efficient micro-lensing of laser-accelerated proton beams by transient electromagnetic (EM) fields in coil targets. In an all-optical principle, high intensity ps-laser pulses are used to charge solid density targets and induce EM target-discharges [1]. The strong transient EM-fields are guided by the target geometry. Such EM-mode propagation along wire targets [2] has already been used for the guiding of a proton beam [3]. Our collaboration aims at a more easily tunable energy-selective collimation and focusing with independent discharge and particle source targets: A sub-mm coil shaped part of the discharge target’s rod produces lensing effects. Protons within an energy range of approximately ±2 MeV, with energies up to 12 MeV, are focused over cm-scale distances.

Experiment

A 50% subdivision of the Petawatt High Energy Laser for Heavy Ion Experiments (PHelix) with 500 fs, 50 J focused at 5 × 1018 W/cm2 into a 10 μm diameter focal spot drives the discharge of a flat 50 μm-thick Cu disc. Targets comprise such a disc as well as a coil of 500 μm diameter. In Capacitor-Coil-Targets (CCT) the disc is part of a capacitor, linked via the coil. Such geometry, shown in figure (1a), allows comparison to previous experiments in the ns-laser pulse regime [4]. With Disc-Coil-Targets (DCT), we conduct a first test of flat targets of simple geometry, depicted in figure (1b).

Main diagnostic is ps-time-resolved imaging of mm-sized areas by proton deflectometry, detected by a a stack of Radiochromic Films (RCF), see figure (1c) and (1d). Proton acceleration from 10 μm thick Au foils is driven by the second 50% subdivision of the PHelix beam, we obtain a proton cutoff energy of 18 MeV.

Two experimental configurations are applied to both target types. A first setup, depicted in figure (1c), and a second setup, depicted in (1d), for field strength determination where the coil’s axis z is perpendicular to the proton beam symmetry axis ⟨⃗v⟩ and a second setup, depicted in (1d), for micro-lensing where ⟨⃗v⟩ ∥ z.

Results

Proton deflectometry reveals the propagation of transient EM-fields following the target geometry with a phase speed of (0.95 ± 0.05) × c. Figure 2 shows results from both CCT and DCT in lensing configuration, with the discharge streaming around the coil over ≈ 25 ps which produces efficient focusing of the protons passing inside it. Up to 12 MeV-protons are collimated over distances of ≈ 5 cm and the beam emittance is reduced by a factor ≈ 3 compared to cases without driving the coil. A closer look on straight conductor sections makes it possible to estimate the E-field component to a peak value of several 10 GV/m.

References

Picosecond-laser driven ultra-fast EM fields propagating along coil targets for proton beam micro-lensing

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Abstract
Efficient micro-lensing of laser-accelerated proton beams by transient electromagnetic (EM) fields in coil targets has been demonstrated [1, 2]. In an all-optical principle, high intensity ps-laser pulses are used to charge solid density targets and induce EM target-discharges [3]. The strong transient EM-fields are guided by the target geometry [4]. A sub-mm coil shaped part of the discharge target’s rod produces lensing effects: protons within an energy range of approximately ±2 MeV, with energies up to 12 MeV, are focused over cm-scale distances. Our collaboration aims at the better understanding of such EM-mode propagation and its pulse shape.

Experiment
A 50% subdivision of the Petawatt High Energy Laser for Heavy Ion Experiments (PHELIX) with 500 fs, 50 fJ focused at 5 × 10¹⁸ W/cm² into a 10 µm diameter focal spot drives the discharge of a flat 50 µm-thick Cu disc. Transient EM-fields propagate with a phase speed of (0.95 ± 0.05) c through the target rod. The discharges follow the target geometry and stream around the coil of 500 µm diameter over ≈ 25 ps, then producing efficient focusing of the protons passing inside it (see figure 1). Up to 12 MeV-protons are collimated over distances of ≈ 5 cm and the beam emittance is reduced by a factor ≈ 3 compared to cases without driving the coil: for example from initially (1.59 ± 0.05) mm mrad to (0.5 ± 0.1) mm mrad for 6.3 MeV protons.

Discussion
Rapid laser-induced target charging leads to a positive potential close to the front surface [3] and a supplementary positive potential at the target rear side [5]. Inside the target, neutralization currents form on a timescale of fs. The positive potential attracts adjacent electrons and propagation of a positive charge distribution starts along the target surface. Using the transverse E-field component around straight conductor sections, a charge-density distribution ρ(F, t) is deduced from deflectometry data and Poisson equation. It can be described as a reduced Cristal Ball Function [6] with a rising edge of ≈ 3 ps and a peak value of several nC/mm. Charge density and displacement current, shown in figure 2, are linked by continuity equation assuming constant phase speed.

Simulation of such an EM-mode propagation with the Particle Field Interaction (PAFIN) code [7] reproduces fairly well the proton deflectometry data, revealing E-fields of the order of the GV/m and B-fields of a few tens of T at the coil center. Comparison of RCF films with synthetic images is shown in figure 1. We find a good agreement with experimental results for a total discharge of ≈ 250 nC.

References
Particle acceleration from levitated targets at Phelix

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**Experiment**

The vast majority of laser ion acceleration experiments relies on thin foils as targets. In the experiment P108 we used isolated spheres with micrometer extent as targets. An electrodynamic trap (Paul Trap) was used to levitate spheres made of copper and plastic (PS). By an electro-optical damping method the trapped particles’ residual motion could be reduced to sub-micron extent and allowed overlapping the laser focus and target reliably. The PHELIX laser delivered 150 J in 500 fs resulting in peak intensities larger than \(10^{20}\) W/cm\(^2\). For enhanced contrast the uOPA front end of PHELIX was used. A magnetic slit spectrometer (covering +\(4^\circ\)) under 0\(^\circ\) served as particle diagnostics. Due to the sub focus dimensions of the target, part of the focal spot is modulated by the target in amplitude and/or phase. The resulting diffraction images were measured via a scatter screen in front of the ion spectrometer. These diffraction images deliver valuable information about the plasma size during the main interaction. In the horizontal plane, at 20\(^\circ\) we used a permanent magnet quadrupole doublet to refocus the accelerated ions. Normal to the laser axis X-ray diagnostics were applied (X-ray spectrometer, X-ray edge source size measurement (for details see progress report of experiment P109)). Reflected light from the target was re-collimated by the parabola and analysed behind a beam line mirror. The setup is shown in Fig. 1.

Fig. 1: Experimental setup.

**Preliminary results**

We were able to reproduce the experimental findings of our previous beam time for 1 µm plastic spheres (experiment P65 in 2013). For 1 µm plastic spheres we again obtained quasi monoenergetic proton spectra with peak energy around 25 MeV and a few MeV bandwidth.

For 2 µm diameter spheres preliminary analysis indicate much higher proton energies of 40-50 MeV. The spectra for even larger targets with 6 µm diameter depended on the hit “quality”. A “full hit” (no transmitted light, i.e. dark scatter screen) delivered a strongly modulated spectrum with about 100% energy spread. A “grazing shot” showed mono-energetic behaviour, indicating the strong influence of pre-plasma expansion on proton acceleration. Copper spheres with 1.5 µm diameter delivered energetic copper ions with a few MeV/u.

The spectrum of the recorded light reflected from a plastic sphere is given in Fig 2. The yellow and green lines depict the expected positions for the second and third harmonic, respectively. The spectrum of the third harmonic is trimmed by the transmission characteristics of the beam line mirror at short wavelengths. The spectra exhibit significant redshift up to 7% of the respective center wavelengths (527 and 351 nm). The observable peaks in the second and third harmonic show the same relative redshift. Transmission screen data indicates that the observed redshift is attributed to the mean electron drift velocity of all target electrons.

We demonstrated the refocusing of energetic copper ions with a set of permanent magnetic quadrupoles. Hereby we observed ion energies which we did not detect in the slit spectrometer.

**Conclusion**

Sub focus sized targets show remarkable source properties such as quasi monoenergetic spectra with large amounts of particles contained in the target being accelerated in forward direction. Due to the finite size of the target the experimental signatures are very clean and without almost any background. This renders levitated targets as an ideal tool for the investigation of laser plasma interactions. All given values are preliminary and currently evaluated in more detail.

Fig. 2: Spectrum of the reflected light
Laser-driven acceleration of deuterium ions from cryogenic targets

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Cryogenic Targets

In the research of laser-driven ion acceleration, a multitude of different materials and designs have been used as the matter sample (target). Gaseous materials pose a special challenge due to their volatility. While gas jet targets are often used, solid targets have a much higher density. Cryogenic targets offer the possibility to use gaseous materials for targets at solid density.

When a target interacts with the laser, not all of it is accelerated as an ion beam. Some material evaporates and some parts remain solid but move as high velocity debris particles. Thus, as opposed to gas jet targets, solid targets carry the risk of damaging optics or other equipment around the interaction point through debris. In addition, vaporized material solidifies again on surfaces at room temperature. As more and more laser systems with repetition rates in the range of Hz are developed, the issue of vaporized target material coating optical elements should be addressed. Cryogenic targets open up the possibility of debris-free solid state targets whose remains can be removed by vacuum pumps.

Deuterium as Target Material

Hydrogen as target material is the only substance that, once ionized, is a pure proton source. Its low triple point temperature of 13.9 K [1] poses a special challenge regarding the needed cooling power.

The hydrogen isotope deuterium can be advantageous due to its higher triple point temperature of 18.7 K. Deuterium ion beams are applicable for e.g. the production of laser-driven neutron beams [2]. In addition, due to their charge-to-mass-ratio of 1:2, deuterium ions can be separated from protons in ion spectrometers, so a distinction of ions regarding their source is possible: Target material (deuterium) ions are discernible from contamination ions such as protons from residual water.

Target Creation

Cryogenic targets are fundamentally different from conventional solid state targets, as they need constant cooling and thus have to be produced and characterized in-situ. A condensation method was developed at the Target Laboratory of the Institute for Nuclear Physics of Technische Universität Darmstadt. Instead of moving through the phase diagram from gaseous directly to solid, a path via the liquid phase was chosen. This entails pressure above the triple point. A setup was developed to combine temperature and pressure control, which was then moved to the target chamber for a beamtime at the PHELIX laser system, conducted in February 2016.

The target is created in an aperture of 1 mm diameter in a cooled copper frame. The required pressure for liquidation is reached by encasing the frame with a motorized growth chamber, which is filled with gas. Liquid covers the aperture and is solidified through cooling below triple point temperature. The growth chamber is then removed to allow access for the laser beam.

As the PHELIX target chamber is larger than the test vacuum chamber at the Target Laboratory and is equipped with more powerful vacuum pumps, we found a difference concerning target survival (see [3]).

Target Characterization

Target thickness characterization, just like target creation, needs to be conducted in-situ. Chromatic-confocal sensors produce axial chromatic aberrations through a set of dispersive lenses. If the sensor is placed in front of a reflective surface, analysis of the wavelength of the reflected image can be processed to the distance between surface and sensor. Thickness values were deduced from distance values to the target surface, under utilization of the geometry of the setup (see [3]).

As the sensors are placed directly in front of the target and thus in the laser beam path, they need to be removed before laser interaction.

Ion Acceleration

Deuterium ions were successfully accelerated during the experiment in 2016 at PHELIX. A detailed publication with results and analysis is in preparation.

References

Resonance spectroscopy with a laser-driven neutron source∗†

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Laser-driven neutron sources have been a topic of intense research and development over the last years. They provide an exponentially decaying energy spectrum with cut-off energies of a few 10 MeV up to over 100 MeV [1,2]. However, there are applications where neutrons with thermal or epithermal energies are preferable. To maximize the neutron yield in this energy range, a moderating material is used to slow down high-energy neutrons. The preferred material should scatter neutrons without absorbing them. Thus, it should contain light elements, like hydrogen or carbon, to maximize the energy loss per collision.

Amongst many other things, neutrons can be used to probe materials. One example of such a measurement is neutron resonance spectroscopy (NRS). This technique utilizes the unique resonance structure in neutron reaction cross sections of elements, which serves as a fingerprint for instance to identify the elemental composition of bulk materials. To investigate the applicability of a laser-driven neutron source for NRS, we conducted an experiment at the Trident laser facility at Los Alamos National Laboratory, USA.

NRS on a static indium sample

During the experimental campaign, thin deuterated polystyrene foils of a few 100 nm were irradiated by the Trident short pulse laser with an energy of 80 J and a pulse length of 600 fs. The laser was focused to intensities above 1020 W/cm2 with a f/1.5 off-axis parabola. The accelerated ions from the target impinged on a 30 × 30 mm cylindrical beryllium catcher (converter) that had a 15 × 15 mm cylindrical hole drilled into it. The hole is directed towards the incoming ion beam. Neutrons that are emitted in the direction of the laser, are scattered back into the catcher to maximize the neutron yield. To moderate the generated neutrons, the beryllium catcher was surrounded by a high density polyethylene (HDPE) block with a length of 13.8 cm and a width of 7 cm. Regarding the setup, there are four main points to be considered:

- Very fast neutrons with energies of several 10 MeV and above (depending on the moderator material and length) are able to pass the moderator without being scattered and thus are not moderated.
- Moderated neutrons change their propagation direction, because they are scattered out to the sides of the moderator.
- The thicker the moderator, the more high energy neutrons get slowed down.
- The moderator dimension in line of sight to the NRS detector is to be kept as small as possible, because the moderator thickness causes a temporal spread of the neutron pulse.

Considering all these conditions, it is advantageous to set up the detector in a sideward direction. The full experimental setup is sketched in figure 1. The neutrons were collimated with a set of boron-treated polyethylene (B-PE) blocks and steel disks. The sample was an indium sheet with 5 mm thickness placed at 1.67 m directly in front of a boron-doped microchannel plate (MCP) detector, which was additionally shielded with 2 mm lead to stop gamma rays from the interaction of neutrons with the indium sample, and B-PE to all sides to avoid detection of scattered neutrons.

With this setup, we were able to measure the 1.45 eV 115In resonance and thus could successfully demonstrate the feasibility of a laser-driven neutron source for NRS.

References

Laser pulse amplification by Stimulated Brillouin Scattering (SBS) and cell radiation damage caused by laser accelerated protons

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Introduction

Strong light pulse amplification by Stimulated Brillouin Scattering (SBS) is a new, prospecting way to realize a laser gain medium that is able to sustain much higher intensities than a solid state body, as commonly used today. Hence, short pulse amplification by SBS could be used to reach extreme laser intensities with a relatively compact and inexpensive laser setup. This will allow accelerating particles to energies that significantly exceed values achieved in present laser particle acceleration.

In 2015, a successful experiment was carried out at the Düsseldorf Arcturus Laser Facility. The Arcturus laser system generates three synchronized beams, with variable pulse durations $\geq$ 25 fs and energies up to 5 J. The two 200 TW beams were used for ionization of the gas target and as a pump whereas the third 3 TW beam line was used as a seed pulse for amplification. The goal of the SBS campaign was to study the SBS process with ultra-short pulses for the first time and find optimal parameters to trigger SBS amplification. In 2016, a second experiment was carried out again at the Arcturus Laser Facility in the so-called strongly coupled regime.

In addition, a study was performed in order to distinguish the biological effectiveness and cellular responses upon irradiation with two types of proton deliveries namely the laser accelerated protons by the Arcturus laser system and conventionally accelerated protons by PTB Braunschweig. High-energy protons induce DNA damage in the form of double stranded and single-stranded DNA breaks (DSB and SSB), DNA base damage and clusters thereof (complex DSB). DNA damage is either induced by direct energy deposition to the 2-deoxyribonucleotide or the bases of DNA (direct effects), or the generation of radicals through water radiolysis, which then interact with DNA (indirect effects). The biological effectiveness of protons is mostly a function of the frequency and complexity of the DNA breaks resulting from direct and indirect effects.

Results

For the SBS experiment, three counter-propagating laser beams, having pulse durations between 30 and 800 femtoseconds and energies up to 800 mJ, were focused onto a gas target. The interaction region was probed by seven diagnostics, that measured energy, frequency and duration of the outcoming laser light. A preliminary analysis of the data shows that a relative amplification of SBS by a factor 6 or more was observed, which is an improvement of factor 4 compared to 2015. In the second study a remarkable difference between the two types of sources of accelerated protons was observed at the same dose level. Cellular nitroxidative stress response is significantly lower after exposure of cells to laser-driven than to conventionally accelerated protons. The results were published in Ref. [1]. The salient finding of our study is that laser accelerated protons (LAP) and conventionally accelerated protons (CAP) have a similar effectiveness to induce DSB. However, LAP exhibited a far lower potential than CAP to induce nitroxidative stress leading to tyrosine-nitration (see Fig. 1). The most likely explanation for this unique property of LAP is the excessively high dose rate. It appears as if redox chemistry changes when dose deposition occurs as an ultra-short pulse, with similar duration as the lifetime of the primary radicals thereby generated (as is the case with LAP).

References

Platform development for laser accelerated particle induced nuclear reaction studies utilizing RC methods*

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Introduction

A team of GSI and LLNL researchers successfully completed a platform development experiment to enable future nuclear science experiments at short-pulsed laser facilities. The experiment is a joint effort between Plasma Physics group led by V. Bagnoud and radiochemists from the Super Heavy Element research group at GSI led by A. Yakushev. This first experiment demonstrated an efficient collection of isotopes produced in nuclear reactions with laser driven MeV proton beams. The proposed experiment was awarded the requested run-time of 20 shifts and 40 shots have been used to demonstrate the isotope collection efficiency and reproducibility.

Experiment

The experiment utilizes laser accelerated proton beams (5 – 20 MeV) in combination with radiochemistry based isotope analysis to study nuclear reactions. In particular, the $^{63}$Cu(p,n)$^{63}$Zn reaction was used to measure the proton activated radioactive $^{63}$Zn via its $\beta^+$ decay and subsequent 511 keV gamma emission identified by its $T_{1/2} = 38$ sec half-life.

The proton beams were produced through the TNSA (Target Normal Sheath Acceleration) mechanism utilizing GSI’s PHELIX laser facility. Laser pulses at 90 J and 500 fs impinging on a thin gold target (the observation of laser accelerated protons has first been reported from experiments at LLNL laser facilities [1,2]). After various tests to identify the spatial, time and energy distribution the accelerated protons were used to activate thin $^{63}$Cu-foils and to measure the level of activation, which was found to be consistent with known milli-barn cross sections for 5 – 20 MeV protons. Due to the divergence of the accelerated protons over about 30 mm, the spot size at the interaction with the target foils is around 10 mm. The spot size of the accelerated protons at the exit of the conversion foil is less than 1 mm due to the small source emittance.

After passing through a thin capton foil as debris shield and a thin Ti-foil as window in the gas filled (in flow mode) target cell inside the main target chamber, the protons interacted with $^{63}$Cu target foils (1-5 stacked foils) inside the target cell. The produced $^{62}$Zn recoil isotopes were stopped in a He/aerosol gas mixture at about 1 bar gas pressure, and transported to a filter through a thin tube. The decay of the $^{62}$Zn was identified by measuring the 511 keV annihilation gammas following the $\beta^+$ decay with a half-life of 38 min (Fig. 1). The reaction $^{63}$Cu(p,n)$^{62}$Zn and gas transport was verified by comparing shots with and without $^{63}$Cu foils and with and without carrier gas. The length of the transport line was varied from a 20 to 1 m distance resulting in a transport efficiency varying between 10 to 60%.

![Figure 1: 511 keV gamma emission following the $\beta^+$-decay of $^{63}$Zn after gas collection utilizing a 1 m (top) and 25 m (bottom) capillary transport line; the decay of the 511 keV gamma line confirms the 38 sec half-life of the $^{62}$Zn isotope.](image)

The presented experiment at PHELIX with laser accelerated MeV protons and a pulse duration of around 1 ps opens up new domains of studies through the detection of short lived isotopes. The experiments provide critical experimental input for future experiments at FAIR (Facility for Antiproton and Ion Research), but also at laser facilities as ELI (Extreme Light Infrastructure) [3,4]. The collaborative experimental research is part of an agreement between LLNL (DOE) and the GSI (BMBF).

References


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Development of a high-resolution x-ray spectrometer for laser-generated hot dense plasma emission *

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In last year’s GSI Scientific report we reported on the generation of ultra-high energy density conditions irradiating tailored micro-pillar arrays at high laser-drive energy [1]. We carried on further experiments to better characterize these extreme plasma conditions. A high-resolution x-ray spectrometer was developed so as to measure the variety of K-shell emission lines providing accurate information about the time evolution of both temperature and density. We report here on the technical design of that instrument.

In our experiments, the target material chosen for spectroscopy purposes is copper. For this element, the K-shell transition lines of interest are emitted between 8000 and 9000 eV, ranging from quasi-neutral species’ Kα (∼8040 eV) to highly-ionized ion emission (8370 and 8680 eV for Heα and Lyα, respectively). Furthermore, many intermediate lines from various charge states and so-called dielectronic configurations can provide accurate estimates for the plasma parameters. In order to differentiate these spectrally-close lines, we need high quality crystals which are able to perform high-resolution diffraction. α-Quartz silicon dioxide (SiO2) crystals are commonly used for this application [2] and are widely characterized [3]. These quartz crystals are available with various lattice cuts, which has to be chosen in accordance with the corresponding Bragg angle. The ideal choice for the spectral window of interest is the lattice 2243 with a 2d-spacing of 2.024 Å. Finally, increasing the signal-to-noise ratio is made possible with spherically curved crystals which focus the spectrum into a thin line.

A commercial crystal (Golem IMS GmbH) is used. It consists of a 12 × 48 mm² quartz crystal (2243) applied onto a concave spherical glass substrate with a radius of curvature of 150 mm. We designed a holder to position the crystal onto the spectrometer board (Fig. 1). The crystal is mounted on a kinematic platform (Thorlabs KM100B/M) equipped with two tip-tilt adjusters for fine alignment of the crystal pointing. The platform is attached to an assembly of two translation stages (OptoSigma TSDH-251S and TSDS-253): one along the x-axis parallel to the spectrometer board in the crystal focus direction and another one along the z-axis for crystal height adjustment. The spectrometer board is a 10 mm-thick Delrin plate which extends in the direction of the central axis of the crystal so as to mark the position of two alignment reference points with the help of pointy metal dowel pins. These two reference points are: (1) the center of the Rowland circle (focus of the crystal at 0°), and (2) the center of curvature. They are located at 75 and 150 mm from the center of the crystal surface, respectively. Finally, the spectrometer board is mounted onto a rotation stage (Newport M-UTR80SA) whose rotation axis passes through the center of the crystal surface in order to maintain the crystal in the right position while adjusting the Bragg angle by turning the plate. The detector, an imaging plate (Fujifilm BAS-SR), is placed in an enclosed aluminium box. An alignment hole (3) is located on the side of the box so that a laser diode shining through it would span the surface of the detector across its long side, thus symbolizing the detector axis. This method is used to finely tune the focus of the spectrum as the geometrical configuration requires that the radiation source and the center of curvature of the crystal are placed on the detector axis.

In the framework of the BMBF project “Development of x-ray spectroscopy for ‘Day One’ plasma physics experiments at FAIR”, we will delve into further designing of similar spectrometers.

References


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Ultra-high energy density conditions produced in free-floating micron-size targets by intense laser irradiation

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In relativistic laser-matter interaction a large fraction of the laser energy is converted to a population of energetic (MeV) electrons. When using “mass-limited” targets with dimensions smaller than the hot electron range the hot electrons are confined to the target by electrostatic sheath fields, leading to the generation of matter at ultra-high energy density (see e.g. [1,2]). While individual mounting of such targets has been realized by e.g. using spider-silk threads, the holding structure constitutes a possible source for return currents and drain for the hot electrons. Avoiding any nearby material, truly mass-limited targets can be realized using droplet-targets formed by the Rayleigh-decay of a liquid jet [3].

In the experiment described here we have made use of a Paul-trap setup, developed at the LMU, Munich [4]. Solid spheres with diameters of order 1 µm were charged up with an ion-gun and held freely floating by the electrostatic fields in the trap. Employing active damping allowed overlapping the few-micron laser spot with the targets.

At the PHELIX laser facility we have subjected micron-sized solid copper spheres to short (~500 fs), energetic (150 J) laser pulses, focused to intensities above 10^20 W/cm^2. We have made use of the uOPA front-end amplifier system, providing laser pulses with ultra-high temporal contrast, to limit heating and expansion of the target before the arrival of the peak of the laser pulse.

Energetic electrons generated in the laser-matter interaction penetrate the solid target, simultaneously heating the material by collisional energy loss and producing inner-shell ionization. The latter is followed by the emission of K-shell radiation which can serve as a diagnostic of the target state. We have developed a highly efficient broadband crystal spectrometer based on a large cylindrical crystal of highly-oriented pyrolytic graphite. The spectral range of the spectrometer from 7.5...12 keV covers the entire copper KL-emission spectrum, from neutral K-alpha up to Ly-alpha from hydrogen-like copper ions. The spectrometer was designed such that the passive detector (Fuji imaging plate) is located outside the target chamber to allow easy exchange of the detector between shots without breaking the vacuum.

The x-ray emission source size was measured by means of a knife-edge setup. A laser-cut gold foil was positioned at a distance of 25 mm from the target and the edge projected onto an image plate detector located at a distance of ~2.5 m outside the target chamber. The resulting magnification of ~100x provided a resolution of approx. 1 µm. With the spot size of the laser focus of 5 µm significantly larger than the targets, the laser pulse energy intercepted by the target amounts to less than 2 J. Nevertheless, due to the small dimensions this results in a laser energy per target volume of 10^9 J/mm^3, at least 2 orders of magnitude larger than previous experiments at kJ short-pulse laser facilities [5].

K-shell emission x-ray spectra show strong emission from the H-like and He-like resonance lines. The complete absence of any emission from lower charge states indicates rapid ionization of the entire target during the time of interaction with the laser pulse. This is supported by particle-in-cell calculations of the laser-target interaction and subsequent target evolution. The simulations show rapid volumetric heating within the first half of the laser pulse to bulk temperatures of 3–4 keV, concomitantly with ionization to charge states exceeding 27+. Atomic kinetic calculations of K-shell emission spectra at these conditions agree well with the measured spectra. The simulations also show the fast hydrodynamic expansion of the target. However, during the few hundred femtoseconds laser pulse duration the rarefaction wave, although at sound speeds of several hundred km/s, has only progressed into the target by a fraction of the target radius.

The x-ray emission source size is measured to be comparable to the initial target size, indicating negligible expansion of the target during peak emission. This could be explained by a rapid relaxation of the hot electron distribution, for example by adiabatic cooling.

Bulk temperatures of several keV are reached while most of the target mass is still at solid density, corresponding to pressures exceeding 10 Gbar. This opens a path towards studying matter at ultra-high energy densities in the laboratory. Upcoming x-ray free electron lasers will be ideally suited to probe structure and dynamic of these highly excited states of matter. At our conditions the high densities and charge states yield a strongly coupled ion system, however with non-degenerate electrons due to the high temperatures.

References

Time-resolved measurement of the relativistic interaction of an ultra-intense laser pulse with sub-micrometer-thick targets


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Motivation

For a better understanding of the interaction between laser and thin foils during experiments at the PHELIX laser and general laser-plasma experiments temporally resolving the interaction dynamics can bring a new insight. Conventional diagnostics are too slow to resolve those interactions, but the method of frequency resolved optical gating (FROG, [1]), could be used as a ultra fast diagnostic. Such a FROG-device has been specifically designed for this type of experiment [2].

Experimental Setup

The experimental setup can be seen in figure 1. The pulse of the PHELIX laser is focused on sub-micrometer-thick CH₂ targets. A part of the pulse is reflected at the critical plasma density, which is picked up and back-collimated by the focusing parabola. After leaking through a mirror, the back-reflected pulse is split up in two parts and sent to the FROG-device, as the main diagnostic, as well as a 1 ω spectrometer. For the measurement of the transmitted part of the pulse a sub aperture of the beam is taken, which is collimated and imaged on to a second FROG-device and a spectrometer. The rest is blocked by a glass-screen which is used to determine the amount of transmitted light.

Preliminary Results

For both reflection- and transmission diagnostic, different modulations of the pulse could be observed, which can be explained by different physical effects. Preliminary results for the spectra can be seen in figure 2. The back-reflected spectrum experiences a larger red shift than the transmitted spectrum. This can be attributed to the doppler-effect of the laser-hole-boring [4], whereas both spectra are broadened due to relativistic self-phase-modulation. [5] A reconstruction of those pulses with a FROG-algorithm [1] can now show the temporal behaviour of the pulses, which is currently ongoing.

Figure 2: Spectra of the transmitted and reflected part of the laserpulse compared to the incoming reference pulse.

For thin targets in the order of a few hundred nanometer a measurement of the transmitted double pulses show a clear retarding effect on the post pulse, which rises up to 500 fs for 200 nm thin targets. This retardation can be used to gain additional information about the pre-expansion of the target before the main pulse arrives. A more detailed analysis of this double pulse setup can be found in [3].

Conclusion

Both the use of FROG as main diagnostic and the double pulse configuration show promising results which will be further evaluated and also used in upcoming experiments.

References


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Development of a FROG for temporal resolution of laser-plasma interactions

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Motivation

During typical relativistic laser-plasma experiments, a complex interaction between the laser and the plasma takes place. For experiments employing thin foils, a part of the pulse can be transmitted and/or reflected by the plasma generated by the interaction. Due to effects such as relativistic self-phase-modulation [1] and doppler-effect [2] the laser pulse is distorted after the interaction. This distorted pulse carries some information on the interaction conditions, and therefore the different effects can then be identified by spectrally and temporally resolving the pulse, which can be achieved by the technique of frequency resolved optical gating (FROG) [3].

Setup

The FROG setup consists of a second-harmonic autocorrelator, an imaging system and a self-constructed 2D-spectrometer with a high dynamic range. Our experimental implementation can be seen in figure 1. The device is designed to resolve the pulse of the PHELIX laser with a pulse duration (FWHM) of 500 fs and a spectral window of 14 nm, as well as pulses with a high spectral broadening of up to 200 nm and slightly stretched pulses up to 2 ps. The self-constructed 2D-spectrometer is able to resolve these ranges with a spectral resolution of up to 0.07 nm and a temporal resolution of up to 15 fs.

Verification

As a verification of the FROG, nearly transform-limited pulses as well as highly linearly chirped pulses, have been measured. With the help of a FROG-algorithm the pulses could be reconstructed temporally and spectrally. The behaviour of the spectral phase for different linear chirps was in good agreement with the expected behaviour. A result for the reconstructed spectrum of two pulses, including the spectral phase, can be seen in figure 2.

Figure 2: Reconstructed spectra for the PHELIX laser for a nearly transform-limited (upper) and strong linear chirped (lower) pulse. The black line shows the spectral intensity and the blue line shows the spectral phase. The light areas indicate the uncertainty given by the algorithm.

Conclusion

The FROG has been implemented at the Petawatt target area of the PHELIX building, where it can be used as a diagnostic for back-reflected and transmitted laser pulses during laser-plasma experiments [4]. Furthermore it can be used to measure the unmodified pulse of the PHELIX-laser with a higher precision than the standard autocorrelators.

References

Generation of keV hot near solid density plasma at high contrast laser-matter-interaction * †

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Laser accelerated electrons play a major role in the process of laser energy transfer into matter. Electron energies can usually be described by a maxwellian-like distribution function with one or more temperatures. In this work, we investigate relativistic laser-matter interaction at high laser contrast and show that a large amount of relatively “slow” keV electrons play a dominant role in the target heating process leading to the creation of a high energy density plasma state. The experiment was carried out at the JETI-40 laser system delivering a high contrast (10^{-6}) frequency doubled (400 nm), 45 fs relativistic (10^{19} W/cm^2; a_0 \approx 1) laser pulse. The 180 – 200 mJ laser pulse was focused to a 5 \mu m spot onto the target (25 \mu m-thick Ti foils) at 45 deg. The diagnostic setup used in the experiment included an X-ray spectrometer with a cylindrically bent HOPG-crystal (2d = 6.71 \AA) ensuring a wide spectral window of 4.4 – 7.9 keV and a spectral resolution of \lambda/\delta\lambda = 1000. For analysis of the bremsstrahlung radiation up to 0.5 MeV a hard X-ray detector (HXRD) based on a filter attenuation method and a Timepix detector operated in the single hit regime were used.

Spectral distribution of the bremsstrahlung radiation of suprathermal electrons traversing the target measured using the Timepix detector is presented in Fig. 1. It can be well approximated by a two temperature electron energy distribution with T_1 = 1.5 \pm 0.2 keV, T_2 = 19.6 \pm 0.1 keV and the corresponding hot electron fraction f_2 of nearly 3%.

Fig. 2 presents a Ti spectrum containing several K-shell transitions: K\alpha- and K\beta-transitions of weakly ionized Ti-atoms, K-shell transitions of intermediate charge states with vacancies in the M- and L-shell (F- up to Be-like ions) and K-shell transitions from one and double excited states in Li- and He-like Ti ions. While the intermediate charge states occur at plasma temperatures of 200 – 300 keV, the Li- and He-like states originate from a hot surface layer with keV temperatures. Both, a large number of low-energy electrons and a small interaction volume lead to high energy density plasmas with keV temperatures and near-solid densities. As shown in [1], the analysis of the K\alpha-profile broadening, that incorporates K-shell transitions of weakly ionized Ti-ions, allows to determine a plasma temperature in “warm” foil regions heated by laser accelerated electrons. In our case, this method leads to temperatures of 20 – 50 eV. The fit of the experimental spectrum in 2 was made using FLYCHK [2] for an optically thin plasma case with a bulk electron temperature of T_1 = 1250 eV. Diagnostics of the electron density was based on the relative intensities of the H\alpha resonance and intercombination (1s^2(1S_0)–1s2p(^3P_1)) transitions.

Figure 1: Bremsstrahlung radiation measured by Timepix.

Figure 2: Measured Ti-spectrum and FLYCHK fit.

According to the fit made for the optical thin case, we obtain n_e = 1.7 \cdot 10^{23} cm^{-3} or 15% of the Ti solid density. Corrections for optical thickness \tau = R/L_{ph} (see FLYCHK) lead to a higher bulk plasma temperature T_1 = 1450 eV and electron density n_e = 2 \cdot 10^{23} cm^{-3}.

References

2D simulation of a hohlraum backlighter for opacity measurements*

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Multidimensional codes, which combine the solution of the fundamental hydrodynamic equations with the spectral transfer equation for thermal radiation and with an accurate scheme for thermal conduction, provide an indispensable tool for the design and the analysis of experiments as well as for the understanding of physical phenomena at high energy density. The radiation-hydrodynamics code RALEF-2D [1] and the equation-of-state / opacity codes FEOS [2] and THERMOS [3] support the research at GSI and at the upcoming FAIR facility. Furthermore, code development is still in progress [4].

In the past, measurements of the heavy-ion stopping in laser-generated dense plasmas at high temperatures at GSI were of crucial importance for the indirect drive scenario of heavy-ion fusion and for the ion-driven fast ignition concept. The corresponding RALEF-2D simulation results [5,6] for the hohlraum X-ray spectra as well as for the plasma column densities were essential for understanding the measurements and for optimization of the experimental setup. Now, current research for planned warm dense matter experiments at GSI and FAIR focuses on the design of diagnostic options, especially of backlighter sources for opacity measurements.

For opacity measurements of expanding laser-heated plasmas an intense “smooth” backlighter option is needed where the spectrum should not be tainted with dominating spectral lines. For such purpose hohlraum targets are a well-suited option. Fig. 1 shows a simulation of a cylindrical gold hohlraum backlighter target with length 0.8 mm and diameter 0.8 mm heated by the short pulse (10 ps pulse duration and 50 J total deposited energy) option of the PHELIX laser at GSI. In contrast to the “old” above mentioned hohlraum simulations for combined laser-ion-beam experiments, the much shorter laser pulse demands for a better and more complicated numerical mesh structure. The figure demonstrates the ablated plasma from the left hohlraum wall by the laser beam and from the right wall by thermal radiation. Both plasma fronts collide and form a hot filament close to the hohlraum center.

Fig. 2 shows the calculated X-ray spectrum as would have been observed through the lower hohlraum hole at three times and in comparison to a Planckian fit for T = 33 eV. At t = 5 ns the spectrum comes close to the Planckian one with a peak maximum of the spectrum at 100-120 eV. At t = 20 ns, both the matter and radiation temperature close to the hohlraum center drop down only by ~ 15% compared to the temperatures at t = 5 ns. This shows that the hohlraum can be used over a long time period with T = 28-33 eV for the opacity measurements.

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Figure 1: Simulated temperature inside a 0.8 mm long gold hohlraum with diameter 0.8 mm at t = 5 ns. The hohlraum is heated by the short pulse of the PHELIX laser. Black lines indicate the boundary of the hohlraum walls and the direction and spot of the laser at t = 0 ns.

Figure 2: Calculated X-ray spectrum as would have been observed through the lower hohlraum hole at t = 2, 3, and 5 ns. The solid line shows a Planckian fit for T = 33 eV.

References

Charge-state equilibration of a carbon beam at 0.65 MeV per nucleon energy in thin solid carbon foils


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We report on measurements of the charge-state distribution of carbon ions at a 0.65 MeV per nucleon energy behind thin solid carbon foils obtained within the experiment U303. In this low-energy range of the beam-target interaction, the collision processes that rule charge-exchange cross sections cannot be described within a first-Born approximation. The prediction of the charge-state distribution is therefore challenging. Moreover, the experimental database is still very incomplete. Here, we use carbon foils with thicknesses in the range 2–10 µg/cm² for which the beam charge is expected to reach equilibrium [1]. By measuring not only equilibrium but also non-equilibrium charge-state distribution, we are able to absolutely determine the electron capture and loss cross sections.

The experimental setup at the Z6 area of the UNILAC accelerator is shown in Fig. 1. The carbon ion beam at an initial energy of 3.7 MeV per nucleon, after passing through a 1 mm diameter pinhole for beam collimation, was decelerated through a 40 µm thick aluminum degrader foil. This resulted in a beam energy of $E_p = 0.65 \pm 0.015$ MeV per nucleon that was measured using the time-of-flight method with a semiconductor detector based on chemical-vapour-deposition (CVD) diamond [2].

The ion slowing down also created a charge-state distribution of the carbon beam. One of the charge states was then selected using a dipole magnet featuring a length of 60 mm and a magnetic field of 1.2 T strength and employed as a probe for the thin carbon foils. The resulting beam charge-state distribution behind the thin foils was split spatially using another dipole with a length of 60 mm and a strength of 0.8 T and subsequently registered on a scintillator screen with the help of a CCD camera. The charge states 5+, 4+ and 3+ of the carbon beam were successively selected as initial charge states for the measurements. Most foils were produced at the Target Laboratory of GSI and

![Figure 1: Setup for the charge-state measurements](image)

![Figure 2: Measured charge-state distributions for the carbon beam at 0.65 MeV per nucleon energy as a function of the thickness of the carbon foil, for an initial beam charge state of 3+, compared with the predictions of the ETACHA code (3+: plain line, 4+: dashed line, 5+: dotted line, 6+: dash-dotted line)](image)

their thickness was measured by means of Raman backscattering spectroscopy at Helmholtzinstitut Jena with a precision of about 3–5 % for each foil. Additional carbon foils of 2–4 µg/cm² thickness were made at the Target Laboratory of Technische Universität München and measured in thickness with a precision of 5 % with the help of a quartz crystal microbalance.

The measured charge-state distributions are compared with the predictions of the ETACHA code [3] that solves rate equations for the projectile ions. The results for the initial charge state 3+ are presented in Fig. 2. A preliminary analysis suggests that, firstly, the ETACHA code over-estimates the electron loss cross section compared to the electron capture cross section and that, secondly, the charge-state equilibrium is reached for foil thicknesses smaller than predicted. A detailed analysis considering the presence of contaminants on the foils surface is in progress.

References


A light gas accelerator for studies on dynamic material properties with PRIOR∗

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At FAIR a novel diagnostic system, the proton microscope (PRIOR), will use high energy protons for radiography [1]. The ion accelerator will be used for accelerating the protons for diagnostics, thus an external driver for dynamic experiments is needed. At the Technische Universität Darmstadt the design and realisation of a two stage light-gas driver for shockwave experiments is ongoing. The parts and the design of the accelerator are shown in figure 1 and 2. The first stage consists of four pistons driven by methane combustion. These pistons compress and heat up Helium in the second stage. The Helium then is supposed to accelerate a sabot carrying a flyer. According to present estimations the two stage device could accelerate 3 g loads up to about 3 km/s. The flyers will shock load different types of targets. The resulting material states should be investigated by a combination of proton radiography and other means.

The basic experiment setup consists of the light-gas accelerator and PRIOR (see Figure 1). A flyer accelerated with the driver impacts into a pusher or directly into a target. The target then is irradiated by the protons so the proton microscope could serve as an in situ diagnostic of the shock wave inside the target. According present estimations the shockwave and proton pulse should be synchronised with accuracy of 50 ns. The ignition of the accelerator will have a quite large jitter of about 1 ms. Also the velocity of the flyer is not precisely predictable. Thus the velocity of the flyer will be measured at the end of the barrel with a light barrier to produce a trigger signal for the kicker magnet. This kind of inverse triggering means that the accelerated protons have to been stored inside the synchrotron up to the arrival of the trigger signal. For a 4 mm target and a shock velocity of 3 km/s the shockwave inside the target will need about 1.3 μs. To get a picture of the shockwave itself the proton beam has to illuminate the target in this time. Typically a beam with 4 bunches in one Proton pulse (0.7 μs) should be used. To adjust the flight time of the projectile after the measurement between 2.5 and 200 μs the flight distance will be adjustable. The possible precision is limited by the distance between the bunches in the synchrotron and the accuracy of the velocity measurement.

The goal of the planned experiments is to investigate material properties under shock wave loading. Also it is possible to study the behaviour of different kind of surfaces. The proton radiography will allow to measure the density distribution inside the shocked materials. Targets for this kind of experiments are currently developed and tested at IPCP RAS.

References

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Investigation on the theta pinch plasma as application for an ion stripper for FAIR*

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Further experiments are planned for March-April due to the technical difficulties in the creation and manufacture of the new circuit.

For the determination of the stripping characteristics, it is vital to estimate the electron density of the pinch plasma time-resolved along the beam line. To cover this task, a Mach-Zehnder-Interferometer has been successfully built and tested at IAP Frankfurt. The interferometer operates with a He/Ne-Laser at 632.8 nm wavelength and is based on a heterodyne measurement method with a frequency shift of 80 MHz between the interfering waves, which defines a time-resolution of 12.5 ns. Figure 1 shows the schematic structure of the interferometer.

The preliminary testing of the interferometer has been done without a plasma source by estimating the angle of a glass wedge in a fall experiment to explore basic functionality. The used wedge is indicated with an angle \( \alpha = (3.883 \pm 0.017)\text{°} \) by the manufacturer, which has been reproduced within the error margin with a deviation of only 0.12% from the optimum value. Additionally, a maximum phase error of 0.08 rad has occurred ordinarily during the testing, what implies a lower limit of the measurable line-integrated electron density of \( 4.48 \cdot 10^{19} \text{m}^{-2} \).

References


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Construction, characterization and optimization of a plasma window based on a cascade arc design for FAIR, Status update*

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Introduction

As described in the last GSI Scientific Report [1], the plasma window is a device designed to shield two different pressure areas such that a particle beam may move unhindered through. Since the last report, a lot of effort has been put into determining the operational characteristics, which include the erosion behavior which links strongly to the estimated lifetime of the window, spectra for information about the plasma characteristics e.g. temperature, density, operational pressure and the electrical conditions for ignition and CW burning of the arc. First results prove that a shielding of the gas flow can be achieved with this setup.

Constructional remarks

With the first stable CW-mode of the window established in November 2016, it became obvious that the used cathode material (Tungsten 80%, Copper 20%) proved to be not temperature resistant enough for this application, see Figure 1. Figure 1 shows two cathodes: one brand new (left) and one after the usage of approx. 15min @45A of discharge current.

To encounter this phenomenon, new cathode tips made from pure tungsten were constructed and are tested. Figure 2 shows a first impression of the increased stability for pure tungsten cathode tips. While the W-Cu tip has already melted, the W tip has held its geometry.

Further efforts proved to be necessary: The sealing (hard solder) of the water cooling channels is heavily attacked during the discharge and loses its stability. Thus, several designs including ceramic isolators between the cooling plates, smaller gaps in between are being tested.

Experimental Data

With the establishment of the CW mode, first spectra were taken to determine an approximation of the plasma temperature. Figure 3 shows such a spectrum, from which an electron temperature of about \( T_e \approx 6900 \, K \) can be calculated. This Temperature is less than expected, which might be explained by the composition of the plasma (Ar, C, W, Cu, H).

Status and outlook

The present setup has shown to be capable of shielding two different pressure areas from another without the usage of solid shielding. While the basic proof of principle is adducted, a lot of work is still ahead, especially to increase the lifetime of the cathodes and the discharge channel, e.g. the cooling plates. Right now, several new designs are being tested which should result in reproducible data and CW mode operation throughout this year.

References

Planetary physics research is an important part of the FAIR High Energy density (HED) physics program. For this purpose, a unique experimental scheme named LAPLAS, has been developed, as shown in Fig. 1. A sample material is enclosed in a High-Z, High-$\rho$ cylindrical shell that is facially irradiated by an intense ion beam with an annular focal spot. Such a scheme leads to a low-entropy compression of the sample that generates super-high densities, ultra-high pressures, but low temperatures. Previously, we simulated compression of hydrogen [1] and water [2] using an intense uranium beam with parameters that will be available at the FAIR. The simulations were done using a 2D hydrodynamic code, BIG2 [3]. This work predicted that using LAPLAS scheme, one can generate the physical conditions similar to those which exist in the interior of hydrogen rich (Jupiter and Saturn) and water rich (Uranus and Neptune) planets respectively.

In the present contribution we report calculations of compression of an Fe sample enclosed in a W shell. Studies of Equation of State, transport and constitutive properties of Fe under extreme conditions is key to understanding the interior structure of the Earth and more massive extra-solar rocky planets, the "Super–Earths". The target length is 4 mm, the radius of the iron sample is 0.2 mm and the outer radius of the W shell is 3 mm. The inner radius of the focal spot ring, $R_i = 0.2$ mm and the outer radius, $R_o = 1.2$ mm while the beam is comprised of 1.5 GeV/u uranium ions. Three different beam intensities including $10^{11}$, $2.5 \times 10^{11}$ and $5 \times 10^{11}$ ions per bunch, respectively, are considered. The bunch length is assumed to be 75 ns.

In Fig. 2 we present the pressure distribution in the target on a length–radius plane at $t = 75$ ns using an intensity of $2.5 \times 10^{11}$ ions per bunch. It is seen that a driving pressure of about 2 Mbar is generated in the W absorption region that drives an inmoving and an outgoing radial shock wave. This is shown in Fig. 3, where we present the corresponding density distribution. The optimum compression leads to a pressure of about 4 Mbar, a temperature of around 6000 K and a density of the order of 15 g/cm$^3$, which are expected Earth core physical conditions. Calculations done using $5 \times 10^{11}$ ions per bunch predict pressures of around 7.5 Mbar, temperatures of the order of 10000 K and densities of about 18 g/cm$^3$. Models predict that these extreme conditions may exist at the core of solid rocky planets, 4 – 5 times more massive than the Earth [4].

References
Generation of High Energy Density Fe sample at FAIR employing LAPLAS scheme using a circular beam focal spot *

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The schematics of the proposed experiment is shown in Fig. 1. The sample material "S" which is Fe in this case, is enclosed in a thick cylindrical shell of W and the target is facially irradiated by the ion beam which has a circular focal spot with a Gaussian transverse intensity distribution. We have carried out hydrodynamic simulations of the target implosion using the 2D code, BIG2 [1] which are reported in this contribution. Previously, we did similar calculations using hydrogen [2] and water [3], respectively, as sample material. It was found in the simulations that although the sample materials were directly heated by the beam, the pressure in the surrounding heated part of the outer high–Z shell was still much higher due to the very high density. Therefore, despite the strong preheating of the sample, the sample material was significantly compressed by the outer pressure. Typically, hydrogen was compressed by a factor 10, while a compression of factor 3 was achieved in case of water. In case of Fe, however, the density is about a factor 2 less than that of W. The pressure in the preheated region becomes too high to achieve any noticeable compression. The sample is strongly heated and becomes a hot high pressure liquid at solid density that is confined by the W shell.

In the present simulations, we have used a wide range of beam parameters including the "Day–One" as well as the maximum design values. Four different values have been considered for the beam intensity including $5 \times 10^{10}$, $10^{11}$, $2.5 \times 10^{11}$ and $5 \times 10^{11}$ ions per bunch, respectively. Three different bunch lengths, namely, 50, 75 and 100 ns, respectively, have been used. Moreover, three different values for the focal spot size characterized with full width at half maximum (FWHM), respectively, have been taken into account.

In Fig. 2 we plot $\rho$, T and P vs radius at L = 2 mm at optimum conditions, $2.5 \times 10^{11}$ ions/bunch, bunch length = 75 ns and FWHM = 1 mm.

Table 1: Optimum physical conditions in Fe, $\tau$ = 75 ns, FWHM = 1 mm, N: intensity, $t_o$: time for optimum conditions, PS: physical state and HL: hot liquid.

<table>
<thead>
<tr>
<th>N (10^{11})</th>
<th>$t_o$(ns)</th>
<th>$\rho$(g/cc)</th>
<th>P(Mbar)</th>
<th>$T(10^4$ K)</th>
<th>PS</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>70</td>
<td>8.8</td>
<td>1.4</td>
<td>1.4</td>
<td>HL</td>
</tr>
<tr>
<td>1</td>
<td>64</td>
<td>8.7</td>
<td>2.3</td>
<td>2.5</td>
<td>HL</td>
</tr>
<tr>
<td>2.5</td>
<td>52</td>
<td>8.5</td>
<td>4.0</td>
<td>5.2</td>
<td>HL</td>
</tr>
<tr>
<td>5</td>
<td>45</td>
<td>8.4</td>
<td>5.7</td>
<td>8.3</td>
<td>HL</td>
</tr>
</tbody>
</table>

It is seen that the physical conditions are fairly uniform along the radius and Fe sample is in a hot liquid state. Table 1 shows optimum sample physical parameters in case of different beam intensities. In all these cases, the Fe has been coverter into a hot liquid state. These samples can be used to study the EOS and transport properties, especially the viscosity of HED liquid Fe.

References


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High energy resolution spectroscopy of the target and projectile X-ray-fluorescence ∗†

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Intense uranium beams, that will be available after commissioning of the new synchrotron SIS100 in Darmstadt, can be used for volumetric heating of any type of material and generation of extreme states of matter with Mbar pressures and some eV of temperature. Investigation of their EOS is one of the main goals of the plasma physics program at FAIR. Diagnostic of such extreme states of matter demands development of new diagnostic methods and instruments, which are capable to operate in an environment with a high level of radiation damage. The precise knowledge of the energy density distribution caused by the U-beam in the target is a very important input parameter for numerical simulations of the hydrodynamic response of the target on deposited energy. Simulations are crucial during the planning of experiments and for the interpretation of obtained experimental data. To investigate the energy density distribution, we propose to use the target and heavy ion beam X-ray fluorescence for imaging of the target expansion and mapping of the heavy ion beam distribution in the interaction region with a high spatial resolution of at least 100 μm. The obtained results can be scaled to high heavy ion energies available at SIS18 and SIS100.

First pilot experiments on measurements and characterization of the heavy ion and target fluorescence using pinholes, X-ray CdTe-diodes and dispersive systems have been carried out in 2016 at the Z6 experimental area which is situated after the UNILAC. A 6.5 MeV/u Au26+-Beam passed through different foil-targets (Al, Cu, Ta) of 6 – 10 μm thickness.

Figure 2: Fluorescence spectrum measured by CdTe-spectrometer.

This experiment demonstrated a big potential of X-ray fluorescence as a diagnostic tool for future FAIR-experiments. We observed intense radiation of ionized target atoms (K-shell transitions in Cu at 8-8.3 keV and L-shell transition in Ta) as well as Doppler shifted L-shell transitions of Au projectiles passing through foils in the photon energy region of 10 keV (see Fig. 2). This radiation can be used for monochromatic (dispersive element) or polychromatic (pin-hole) X-ray mapping of the ion beam intensity distribution in the interaction region. Using data obtained by means of CdTe X-ray spectrometer and a faraday cup, we could estimate the number of Au-Lα photons per 1 C of the Au-charge passing through Al, Cu and Ta foils and per 1 μm target-thickness in 4π. This number allows us to conclude that 10-100 fold amplification of the signal is required in order to apply this method for U-beam intensities between 1011 – 5 · 1011 particles/pulse. 2D X-ray pinhole image of the Cu-grating excited by Au-ion beam, obtained with 200 μm spatial resolution, is the first promising result that can be improved in future experimental campaigns.
To study the affect of an accidental release of one FCC beam on equipment and accelerator components, we carried out simulations of the full impact of the beam on a cylindrical Cu target that has a radius of 2 cm and is 5 m long. The beam is comprised of 10600 bunches of 40 TeV protons with a bunch intensity of $10^{11}$ particles. Bunch length is 0.5 ns and bunch separation is 25 ns. The focal spot has a $\sigma = 0.2$ mm. The simulations have been done using an energy deposition code, FLUKA and a 2D hydrodynamic code, BIG2, iteratively. The beam is transversely incident of one face of the cylinder so that the beam and the target axis coincide. In Fig. 1 we present isolines of temperature at $t = 1200$ ns [48 bunches delivered] in the target provided by the BIG2 code. It is seen that the maximum temperature at the axis is $1.2 \times 10^5$ K. Fig. 2 shows that the corresponding pressure has a maximum value of about 46 GPa. The high pressure generates an outgoing radial shock wave that leads to significant density depletion at and around the axis. As a result of this, the protons that are delivered in the subsequent bunches penetrate deeper in the target, which makes the range of the protons and their shower much longer than the static range, which is called, "the hydrodynamic tunneling effect". For example, according to our present calculations, the static range of a single 40 TeV proton and its shower is about 1.5 m in Cu, but the full FCC beam will penetrate up to 295 m in the target [1].

In Fig. 3, we plot profiles of density, temperature and pressure along the target axis. It is seen that the material that lies between $L = 0 - 1$ m is converted into a weekly ionized plasma, whereas material between $L = 1 - 3$ m is in liquid state. This shows that a large part of the target has been converted into different phases of HED matter including Warm Dense Matter and strongly coupled plasma. One may conclude that issues of HED matter are inherent to every powerful accelerator that generates intense ion beams.

References


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Feasibility study of a water beamdump for the 50 TeV FCC proton beam

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After the unprecedented success with the Large Hadron Collider (LHC), a design study is being conducted to assess the feasibility of a much more powerful hadron collider named, Future Circular Collider (FCC). According to this design study, the FCC will be installed in a circular tunnel with a circumference of 100 km and will accelerate two counter rotating proton beams with a particle energy of 50 TeV. Each beam is comprised of 10600 proton bunches while the intensity is $10^{11}$ protons per bunch. Each bunch is 0.5 ns long and the bunch separation is 25 ns.

Safe disposal of the beam after experiment is a very important part of the operation. For the LHC, a carbon beamdump is being used. The beam is diluted before it reaches the beamdump and the bunches are delivered on an extended spiral shaped path so that the energy is distributed over a large volume of the beamdump to avoid material damage. For the FCC beam, such a scheme will require 20 m long spiral path for the bunches, which is very challenging. An alternate beamdump design that uses ordinary water to absorb the beam energy, has been studied with the help of numerical simulations done employing the energy deposition code, FLUKA and a 2D hydrodynamic code, BIG2, iteratively. First results are reported in this contribution.

**Figure 1:** Target geometry.

It is a Cu pipe with an inner radius of 15 cm, an outer radius of 17 cm and is 14 m long (Fig. 1). It is filled with ordinary water and the beam is incident on the left face of the cylinder along its axis. The focal spot is circular with a Gaussian transverse intensity distribution with $\sigma = 0.4$ mm.

**Figure 2:** $\rho$ vs axis at different times.

In Fig. 2 we present $\rho$ along the target axis at different times. It is seen that a density depletion front propagates towards the right which is due to the outmoving radial shock wave. This leads to the hydrodynamic tunneling of the primary beam and the shower [1]. In table I we present the position and the propagation speed of the depletion front at different times. It is seen that the speed decreases with time and achieves a constant value of about $4.80 \times 10^6$ m/s. Since the duration of the bunch train is 265 $\mu$s, the total penetration distance of the beam and the shower will be about 1.3 km, which should be the length of the beamdump. It is interesting to note that the static range of a single 50 TeV proton and its shower is about 7 m in water.

**Table 1:** Position, $x$, Distance traveled in 100 ns, $\Delta x$ and speed, $v$ of depletion front

<table>
<thead>
<tr>
<th>Time (ns)</th>
<th>$x$ (cm)</th>
<th>$\Delta x$ (cm)</th>
<th>$v$ (m/s)</th>
</tr>
</thead>
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<tr>
<td>700</td>
<td>713.00</td>
<td>56.7</td>
<td>$5.67 \times 10^6$</td>
</tr>
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<td>769.70</td>
<td>53.1</td>
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<td>900</td>
<td>822.80</td>
<td>49.8</td>
<td>$4.79 \times 10^6$</td>
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<td>1000</td>
<td>872.6</td>
<td>47.9</td>
<td>$4.82 \times 10^6$</td>
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<td>1300</td>
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**References**