INVESTIGATION
OF PLASMA-BASED OPTICAL
PARAMETRIC AMPLIFIERS AND HIGH
POWER SEED PULSE GENERATION FOR
STIMULATED RAMAN
BACKSCATTERING EXPERIMENTS

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Abstract

Laser pulse intensities have not seen a significant jump in the past ~20 years whereas recent increases in focal intensities are mostly due to reduced wavefront aberrations and subsequent improved focusability. The last paradigm boosting output intensities of laser systems was the invention of the chirped pulse amplification (CPA) scheme in the 90s. In CPA output intensities are limited by the damage thresholds of the solid state optical gratings compressing the amplified pulses. Plasma-based optical parametric amplification is a promising concept to circumvent such limitations. Stimulated Raman backscattering (SRBS) or strongly coupled stimulated Brillouin backscattering (sc-SBBS) are three-wave interactions theoretically capable of generating ultrahigh intensity laser pulses. Surpassing the present intensity limits will be beneficial for various applications (e.g. particle acceleration, fast ignition fusion), open new fields of research (e.g. relativistic ion plasmas) or allow for the validation of present day theory (e.g. nonlinear quantum electrodynamic). Pulse amplification by plasma-based parametric instabilities can also help to build more compact and cost efficient laser systems. However, experimentally acquired results do not show energy transfer efficiencies proposed by theoretical publications. Continuous effort is required to advance plasma-based optical parametric amplification in order to make it a viable scheme for the next generation of ultrahigh intensity lasers.

In this work, experimental results for parametric backscatter amplification in sub-quarter-critical plasmas and the generation of SRBS seed pulses by nonlinear frequency conversion of Bessel beams propagating in gas cells are presented. In the optical parametric amplification campaign for joule-level pump pulses, record amplified output energies exceeding best current results by more than 30% are attained (while conserving high quality spatial profiles and exhibiting short pulse durations around 100 fs). It is suggested that the comparable high energy gains are due to the sc-SBBS instability which is experimentally as well as theoretically scarcely explored thus far.

In a separate campaign self-phase modulated, ultrashort seed pulses are amplified in well-defined plasma density gradients. The sign of the gradient is found to have a significant impact on SRBS efficiency as well as spontaneous Raman backscattering from plasma noise. Strong mitigation of
aforementioned processes are found when increasing the detuning growth rate in backscatter direction by the density gradient. For the gradient partly compensating for the pump chirp a superior bandwidth is achieved compared to flat-top target profiles. One-dimensional PIC simulations are in good agreement with the obtained results and indicate linear amplification in the strong wavebreaking regime of SRBS.

Efficient SRBS at high pump amplitudes and plasma densities requires ultrashort, energetic, redshifted seed pulses. Generation of SRBS seed pulses by (molecular) transient stimulated Raman scattering (SRS) utilizing long caustic focusing in Raman active gases is shown. Required frequency shifts can be achieved by single- or multi-order SRS of the first vibrational mode of the corresponding gas molecules. Frequency shifted pulses are spatially separated from unwanted frequency components (fundamental or anti-Stokes), exhibit a high quality beam profile, smooth spectral curves and experience an almost ten-fold temporal compression. Compared to similar shifting methods (e.g. Raman crystals) orders of magnitude higher output powers and higher output energies are attained which may enable operation in the coherent wavebreaking regime of SRBS in future experiments.
Zusammenfassung


Bild 1: Historische Entwicklung der Maximalintensitäten die im Fokus erreicht werden.

Plasmen haben keine Zerstörschwelle und eignen sich daher als Verstärkermedium für die Erzeugung ultraintensiver Laserpulse. Praktikable plasmabasierte Verstärkungsmechanismen sind die stimulierten Raman-Rückstreuung (SRBS) und die stark gekoppelte stimulirte Brillouin-Rückstreuung (sc-SBBS). Bei beiden parametrisch optischen Verstärkungsprozessen wird ein energiereicher, langer Pumppuls mit einem gegenläufigen, kurzen Seedpuls im Plasma überlagert. Die erzeugte Schwebung kann durch ponderomotive Kräfte eine Dichtemodulation der Elektronen im Plasma erzeugen die sich als Elektron-Plasmawelle ausbreitet (SRBS), bei höheren Pumpintensitäten und Plasmadichten kann alternativ eine quasi-Ionenwelle erzeugt werden (sc-SBBS). Der Pumppuls kann an der jeweiligen Plasmawelle in Seedrichtung rückgestreut und dabei zeitlich komprimiert werden. Für beide Drei-Wellen-Prozesse gelten Energie- und Impulserhaltung die durch $\omega_0 = \omega_1 + \omega_{pl}$ und $k_0 = k_1 + k_{pl}$ ausgedrückt werden können, wobei $\omega$ die Kreisfrequenz und $k$ der Wellenvektor ist, die Indizes $0, 1, \text{und } pl$ stehen für Pump, Seed und Plasma. Elektronen-Plasmawellen haben signifikante Frequenzen (im Vergleich zu Ionenwellen), so dass für SRBS eine Frequenzverschiebung des Seedpulses nötig ist.

Aus den Manley-Rowe-Relationen folgt, dass die Effizienz für den Energietransfer von Pump zu Seed durch $1 - \frac{\omega_{pl}}{\omega_0}$ gegeben ist was in theoretischen Obergrenzen von $\geq 90\%$ resultiert, wobei die restliche Energie zur Plasmawelle transferiert wird. Experimentell wurden bisher jedoch nur Effizienzen von wenigen Prozent und Energien im unteren Millijoulebereich erreicht. Mögliche Sättigungsmechanismen sind Wellenbrechen der Plasmawelle, Teilcheneinfang, Landau-Dämpfung, Modulations-Instabilitäten, Plasmawellenfilamentierung, optische Chirps und Raman/Brillouin Vor- und Rückstreuung des verstärkten Seedpulses bzw. bevor der Pulsüberlapp etabliert ist.

In dieser Arbeit werden Ergebnisse für SRBS in exakt definierten Dichtegradienten präsentiert. Dabei werden am Jeti40 Titan-Saphir-Laser (IOQ, Jena) ultrakurze Seedpulse (~18 fs) durch Selbstphasenmodulation in einer gasgefüllten Hohlkernfaser und nachfolgender Kompression durch Chirp-Spiegel generiert und mit $10^{15}$ Wcm$^{-2}$ Pumppulsen im Plasma überlagert. Vorherige experimentelle Ergebnisse zur stimulierten Raman-Rückstreuung in Dichtegradienten wurden durch Mehrfachionisation des Targetgases oder durch den schrägen Einschluss in vorgefertigte

1D PIC Simulationen für den negativen Plasmagradienten sind in guter Übereinstimmung mit den gemessenen Spektren im entsprechenden Parameterbereich und zeigen eine Pulsdauer des verstärkten, führenden Hauptpulses von ~75 fs (Bild 4). Etwa 90% der Energie befinden sich im ersten Hauptpuls was für Verstärkung unter starkem Wellenbrechen spricht.

In der Strahlzeit am Gemini Ti:Sa-Laser (RAL, Didcot) werden kurze, millijoule Seedpulse mit hochenergetischen Pumppulsen (bis zu 3.5 J, $\sim 10^{16}$ Wcm$^{-2}$) in ebenen Elektron-Dichteprofilen überlagert. Die Frequenzverschiebung zwischen Pump und Seed wird durch eine räumliche Abtrennung der hochfrequenten Anteile des Laserspektrums im Seed-Kompressor realisiert. Der Versatz der Zentralwellenlängen der beiden Pulse beträgt dabei nur rund 10 nm. Dabei werden Energien erzielt, die den aktuellen Rekord für plasmabasierte parametrisch optische Verstärker um etwa 35% übertreffen. Durch die kurzen Pulsdauern der verstärkten Seedpulse (oberer zweistelliger fs-Bereich) wird außerdem der Rekord für die höchste Ausgangsleistung um beinahe eine Größenordnung verbessert (Bild 5). Der spektrale Intensitätszuwachs erfolgt selbst bei vergleichsweise hohen Plasmadichten ($n_e \approx 0.01n_c$) fast ausschließlich im Bereich des ursprünglichen Seedspektrums und ist extrem robust gegenüber Veränderungen der Plasmadichte oder des relativen Pulsdelays. 1D PIC Simulationen mit unendlichen Ionenmassen können Ausgangsspektren und Pulsdauern bei o.g. Plasmadichten nicht reproduzieren. Zusätzlich liegt der Parameterbereich des Experiments theoretisch über der Schwelle für sc-SBBS jedoch noch unter der Schwelle für die superradiante Ramanverstärkung die eine Verstärkung über einen extrem
breitbandigen Bereich ermöglicht. Es wird daher argumentiert, dass Verstärkung durch stark gekoppelte stimulierter Brillouin-Rückstreuung erreicht wurde bei der niederfrequente quasi-Ionenwellen für den Energieaustausch zwischen Pump und Seed sorgen.

Wie zu Beginn erwähnt werden für die stimulierte Raman-Rückstreuung im Plasma Seedpulse benötigt die frequenzangepasst im Bezug zur Plasma- und Pumpfrequenz sind ($\omega_1 = \omega_0 - \omega_{\text{pl}}$). Für typische Plasmadichten und Laserfrequenzen entspricht das einem Wellenlängenversatz > 30 nm.

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Chapter 1

Introduction: The quest for ultrahigh intensity laser pulses beyond current limits

Since the first demonstration of light amplification by stimulated emission of radiation in 1960, the laser has found its way not only into research facilities around the globe but is ubiquitous in industry, medicine and civil society. Scientific applications include spectrometry [1], interferometry [2], plasma-based particle accelerators [3], inertial confinement fusion [4, 5], X-ray lasers [6] and fundamental studies in high energy density physics. Common industrial and medical applications are material processing, sensing, fiber-optic communication, optical data storage and soft-/hard-tissue or eyesight surgery.

Some of the aforementioned applications rely on ultrahigh intensities that can be reached by short laser pulses that are focused onto small areas. Surpassing present intensity limits (~$10^{22}$ Wcm$^{-2}$) will be beneficial for numerous applications such as GeV proton acceleration [17], $\gamma\gamma$-collider [18], superhot matter [19, 20], fast ignition fusion [21] and generally advance experimental capabilities to the frontiers of knowledge.

The demand for more compact and intense laser in order to open up new fields of research or applications is unbroken. For ultrahigh intensities new regimes can be entered, allowing experimental verification of present day theory including nonlinear quantum electrodynamic.
While the attainable laser peak powers have dramatically increased by techniques like Q-switching, mode-locking and the invention of the chirped pulse amplification in 1985, no significant enhancements were achieved in recent years (Figs. 1.1 and 1.2). In CPA, the pulse is temporally stretched to keep the intensity in the amplifier medium below the damage threshold, after amplification the pulse is temporally compressed by solid state gratings to reach up to petawatt output powers [7]. Therefore power is restricted by the damage threshold of the gratings which are currently (depending on pulse duration and wavelength) between $0.3 - 4 \text{ Jcm}^{-2}$. Meter scale gratings and corresponding dielectric mirrors used in PW laser facilities are expensive (six-digit euro figures solely for gratings) and wavefront aberrations (limiting the focusability) are more difficult to control in large-diameter optics. Thus upscaling the beam diameter in order to reach higher output powers would not only mean an upsurge in costs but might also exceed the limits of current technology in terms of grating manufacturing.

An alternative scheme is the combination of CPA with optical parametric amplifiers (OPAs) [8]. Crystal materials exhibiting $\chi^{(2)}$ nonlinearities can amplify a signal beam in presence of a higher frequency pump beam by depleting pump photons into lower frequency signal and idler photons ($\omega_{\text{pump}} = \omega_{\text{signal}} + \omega_{\text{idler}}$). The main advantage to classical laser amplifiers are broad gain spectra that support pulse lengths of a few femtoseconds [9], high gains per unit length and the absence of heat generation which allows high power outputs. Still, the amplifier medium has a specific damage
threshold and compression of extreme broadband pulses is not possible because of grating reflectivity restrictions. It is apparent that there is a need for a new method to surpass the constraints of CPA or OPCPA schemes.

Contrary to solid state optics, plasma does not have a damage threshold. One possible plasma-based amplifier scheme is based on stimulated Raman backscattering (SRBS). It was first proposed in a theoretical work in 1998 [11]. Here a long high power pump pulse overlaps in a plasma with a counterpropagating, frequency shifted, short, comparably low energy seed pulse. The beat wave induces a disturbance in the electron distribution leading to an electron plasma wave (EPW, resp. Langmuir wave) which acts as an optical grating. A portion of the pump is backscattered into the seed pulse and at the same time temporally compressed (in the nonlinear or superradiant regime). It has been shown that this scheme could, in principle, provide amplification and compression to pulse powers in the multi-petawatt regime [10].

The competing plasma amplifier concept is based on stimulated Brillouin backscattering (SBBS) [89, 90]. The aforementioned pulse characteristics are similar. In the backscattering scheme of SBBS a pump and counterpropagating short seed pulse overlap in a plasma. In contrast to SRBS a broadband seed pulse (as obtained from Ti:sapphire systems) practically does not require a frequency shift since the three-wave interaction is coupled to an ion acoustic wave (IAW) which has a much lower frequency compared to the EPW \((m_i \gg m_e)\). The pump is then backscattered at the ion density fluctuation into the seed and temporally compressed. Both schemes, stimulated Raman- and Brillouin backscattering (SBBS), have several characteristics that differentiate them from one another and the preferential process depends on the respective setups and or requirements on the output pulse parameters.

The theoretical efficiencies of the two parametric instabilities are paramount. Manley-Rowe relations dictate that the maximum energy transferred to the seed is given by \(1 - \frac{\omega_{pl}}{\omega_0} \) [36, 37] with \(\omega_{pl}\) the frequency of the EPW, respectively the IAW. For typical plasma densities used in SRBS
(SBBS) experiments this implies that $90 - 95\% (> 99.9\%)$ of the pump energy can be backscattered while the remaining energy is transferred to the corresponding plasma wave.

Most successful SRBS experiments were done at Ti:sapphire lasers and show few percent conversion efficiencies at output energies of a few millijoules (Fig. 1.3). Aforesaid values are far from values predicted by theory and simulations. Effects such as wave breaking, Landau damping, detuning and Raman forward scattering (RFS) or premature Raman backscattering require thorough investigations in order to conceive the discrepancy between theoretical calculations and experimental results and to develop the SRBS instability into a viable tool for future laser systems.

<table>
<thead>
<tr>
<th>Year</th>
<th>Author</th>
<th>$E_{\text{pump}}$ [mJ]</th>
<th>$\tau_{\text{pump}}$ [ps]</th>
<th>$E_{\text{seed}}$ [$\mu$J]</th>
<th>$\tau_{\text{seed}}$ [fs]</th>
<th>$n_e$ [$10^{18}$ cm$^{-3}$]</th>
<th>Eff. [%]</th>
<th>$E_{\text{gain}}$ [mJ]</th>
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<td>M. Dreher et al.</td>
<td>140</td>
<td>3.5</td>
<td>60</td>
<td>80</td>
<td>3</td>
<td>0.67</td>
<td>0.94</td>
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<td>2005</td>
<td>W. Cheng et al.</td>
<td>40</td>
<td>10</td>
<td>7.5</td>
<td>550</td>
<td>11</td>
<td>1.1</td>
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<td>2007</td>
<td>J. Ren et al.</td>
<td>82</td>
<td>20</td>
<td>16</td>
<td>500</td>
<td>13</td>
<td>3.9</td>
<td>3.2</td>
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<td>2007</td>
<td>J. Ren et al.</td>
<td>56</td>
<td>20</td>
<td>3300</td>
<td>90</td>
<td>13</td>
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<td>2010</td>
<td>L. Lancia et al.</td>
<td>2000</td>
<td>3.5</td>
<td>15000</td>
<td>400</td>
<td>100</td>
<td>2.2</td>
<td>45</td>
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<td>2012</td>
<td>D. Turnbull et al.</td>
<td>300</td>
<td>25</td>
<td>100</td>
<td>200</td>
<td>15</td>
<td>4</td>
<td>12</td>
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<td>2014</td>
<td>E. Guillaume et al.</td>
<td>675</td>
<td>15</td>
<td>477</td>
<td>1000</td>
<td>170</td>
<td>2.5</td>
<td>17</td>
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<td>2015</td>
<td>X. Yang et al.</td>
<td>880</td>
<td>250</td>
<td>560</td>
<td>270</td>
<td>1</td>
<td>0.4</td>
<td>3.5</td>
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<td>2015</td>
<td>L. Lancia et al.</td>
<td>6000</td>
<td>4</td>
<td>4000</td>
<td>700</td>
<td>100</td>
<td>0.5</td>
<td>30</td>
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<td>SBBS @ Astra Gemini</td>
<td>3200</td>
<td>6.5</td>
<td>15000</td>
<td>70 - 150</td>
<td>16</td>
<td>1.9</td>
<td>62</td>
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<td>SRBS @ Jeti40</td>
<td>175</td>
<td>3.6</td>
<td>170</td>
<td>&lt; 20</td>
<td>30</td>
<td>≥0.8</td>
<td>≥1.5</td>
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Figure 1.3: Table of results for various plasma-based parametric amplifiers. Green shaded lines show results of SBBS, blue shaded lines results for SRBS. In the two last lines exhibit pulse parameters and results obtained in this work.

SBBS was discarded as a viable plasma amplifier scheme for the longest time because of the assumption that pulses shorter than the ion acoustic timescale $\frac{\lambda}{c_s} \approx 10^{-11} - 10^{-12}s$ (with $c_s$, ion sound speed) cannot be amplified, therefore only allowing for orders of magnitude lower output powers compared to SRBS. However, in the regime of strongly coupled Brillouin scattering, first theoretically proposed in 2006 [91], the compression limit of the seed pulse is given by the inverse growth rate. Subsequent pulse durations can be in the order of $\gamma_{SBBS}^{-1} \approx 10^{-13} - 10^{-14}s$. First experimental results on strongly coupled SBBS (sc-SBBS) were published in 2010 [92] followed by E. Guillaume et al. in 2014 [93] and equally show conversion efficiencies of only a few percent. Limiting factors for the sc-SBBS process are filamentation, Raman forward scattering of the seed, ion trapping and ion wavebreaking.
As can be seen, experimental studies on sc-SBBS are extremely rare. Besides being longer in the focus of interest as a viable plasma-based parametric amplification process, experimental studies on SRBS are equally sparse compared to their theoretical counterparts. Setups for SBBS and SRBS are sophisticated and extensive, available beamtimes at larger laser facilities are often limited and not adequate for setting up the experiment and subsequent data collection.

A part of this work presents the results for what is suggested to be strongly coupled stimulated Brillouin backscattering of picosecond (6.5 – 13 ps), joule-level (0.6 – 3.6 J) pump pulses overlapping with femtosecond (~150 fs), multi-millijoule seed pulses in underdense plasmas \( \frac{n_e}{n_c} \leq 0.009 \). The experiment is performed at the Astra Gemini Ti:sa laser system (CLF). The present record for energy amplification (see Fig. 1.3) is surpassed with an energy gain of ~62 mJ while the output power is increased by a factor of ~5. Effects of relative pulse delays, pump pulse parameters and target densities on the spectral characteristics and energy gain are studied.

Also, first time investigations of stimulated Raman backscattering in well-defined, tailored plasma gradients performed at the Jeti40 Ti:sa laser (HIJ) are presented. Pulse energy and duration is 175 mJ (170 µJ) and 3.6 ps (~18 fs) for the pump (seed) pulse while target densities reach up to \( \frac{n_e}{n_c} = 0.02 \). Acquired results can be directly compared to a flat-top density target (which are commonly used in such experiments). This should lead to a better understanding of the interplay between pump chirp and density gradients, respectively the effects of detuning on the SRBS amplification process.

As mentioned, SRBS experiments require a frequency shifted seed pulse. Common examples of seed pulse generation are Raman crystals (e.g. Barium nitrate or KGW) [12], (non-collinear) OPA [13, 14], self-phase modulation (SPM) [15, 16] or utilizing the low frequency part of the fundamental laser spectrum. Here a new method for generating high quality, multi-millijoule, frequency shifted seed pulses for SRBS experiments by Bessel beam pumping of molecular gases is proposed. Experimental results from two campaigns at the Jeti40 and PHELIX Nd:glass laser (GSI) show orders of magnitude higher output powers compared to alternative shifting solutions. At the same time output pulses are temporally compressed (to the femtosecond range) and spatially separated from the unsuitable frequencies removing the need for additional compressor setups or spectral filtering while input pulse durations are viable for SRBS pumping at the same time.

This work is generally structured as follows, chapter 2 and 3 cover the theoretical descriptions of the plasma-based parametric amplification processes (SRBS, SBBS) and the generation of frequency shifted seed pulses. The corresponding experimental results are then presented in the following chapters 4, 5 and 6.
More specific, chapter 2 is dedicated to the explanation of stimulated Raman- and Brillouin backscattering in plasmas. An introduction into laser ionization mechanisms, collective plasma effects and parametric instabilities is followed by a description of the SRBS and SBBS process and their competing instabilities as well as other deleterious effects that can reduce efficiency of plasma-based amplifiers. Chapter 2 directly relates to chapters 4 and 5. Where the former presents the amplification of seed pulses by strongly coupled stimulated Brillouin backscattering and the latter the amplification in flat-top and tailored density gradients by stimulated Raman backscattering.

Chapter 3 depicts the theory related to the generation of frequency shifted pulses required for SRBS experiments. It includes the characterization of Bessel beams and Raman scattering in gases which is coupled to the vibrational and rotational modes of the corresponding molecules (as opposed to the Raman scattering in plasmas which couples to the electron plasma wave) and gives a short introduction into the effect of self-phase modulation. Chapter 3 should therefore lay foundation for the comprehension of the seed generation at the Jeti40 SRBS campaign (chapt. 5.2.1) as well as the proposed seed generation scheme by Bessel pumping of molecular gases (chapt. 6).

Chapter 7 concludes the thesis with a summary and an outlook to the future of plasma-based parametric amplifiers as an addition to the CPA scheme in order to reach ultrahigh intensities.
Chapter 2

Plasma-based optical parametric amplification

Plasma amplifiers based on parametric instabilities have been proposed since the 1980s. Plasma as an amplifier medium is advantageous compared to solid state amplifiers since its lack of a damage threshold. There are two different parametric instabilities that occur in undercritical plasmas that can be utilized for backward amplification of laser pulses, namely stimulated Raman backscattering (SRBS) and stimulated Brillouin backscattering (SBBS).

Both instabilities can be described by coupled equations for a three-wave interaction. The three waves include two electromagnetic light waves, so called pump and seed pulse, and a plasma wave. Under optimum conditions a significant part of the high energy pump pulse is backscattered into the seed pulse and compressed in time. As a result higher output powers compared to the incoming pump pulse can theoretically be obtained.

The chapter starts with laser-induced ionization processes since in this work pump pulses are utilized as ionizing pulses at the same time. The front of pump pulse generates the plasma in the gas jet target so the remaining part of the pump can interact with the counterpropagating seed pulse inside the plasma. The beat wave can trigger electron (EPW) or ion acoustic (IAW) plasma waves which are discussed in the following paragraph. Dispersion relations for different waves in plasmas are introduced. Thereafter parametric processes in plasma specifically SRBS and SBBS are treated. Different amplification regimes of the two instabilities are depicted, whereas in case of SBBS the focus is laid on the strongly coupled regime. Different mechanisms that can reduce the efficiency of the amplification processes are discussed and evaluated for their significance.
2.1 Laser-induced ionization mechanisms

Plasma is a quasi-neutral gas consisting of neutral and charged particles exhibiting collective behavior. It is possible to generate plasmas by laser-induced ionization of atoms. Classically an atom can be ionized if the photon energy is at least equal to the binding energy of the electron (photoionization). This is not the case for the near-infrared laser photons generated at Ti:sapphire or ND:glass systems (e.g. 800 nm \(\approx 1.55\) eV). For short, intense laser pulses ionization is caused by multiphoton / above threshold, tunnel and over-the-barrier ionization.

For multiphoton ionization an electron successively absorbs photons until the Coulomb potential is overcome. Tunnel ionization occurs for strong electric fields that are able to distort the potential barrier of the atom. Hence, the probability for an electron to tunnel through the barrier is increased significantly. For even stronger fields the potential barrier is further depressed until its peak reaches the binding energy of an electron, which is then able to freely escape from the atom.

The prevalent mechanism depends on the intensity and frequency of the radiation. The Keldysh parameter is an indicator if the ionization process occurs in the regime of multiphoton (\(\gamma \gg 1\)) or tunnel ionization (\(\gamma \ll 1\)) [22]

\[
\gamma = \frac{\omega L}{eE} \sqrt{2I_p m_e}.
\]

With \(\omega L\) and \(E\) the angular frequency and electric field amplitude of the laser, \(I_p\) ionization potential and \(m_e\) electron mass. For a typical pump intensity and working gas in the SRBS experiments \(\gamma < 1\) so tunnel ionization is expected as the prevalent ionization mechanism.

The ionization process and the resulting plasma influence the laser pulse propagation. Energy losses due to inverse Bremsstrahlung and spectral blue shifting appear. The blueshift caused by rapid creation of plasma electrons can be calculated as [23]

\[
\Delta \omega = -\frac{\omega L}{c} \int \frac{\partial n}{\partial t}(l) dl
\]

with \(n\) refractive index and \(l\) interaction length. Energy losses and spectral blue shifting is seen for intense pulses ionizing the gas jet (chapt. 4). This effect must be taken into account since the initial spectrum and energy might not be similar to the actual values after traversing a certain distance in the target.
2.2 Plasma oscillations, plasma waves and dispersion relations

Assuming that the ions in plasmas are an immobile background ensuring quasi-neutrality, the laser pulse solely interacts with the plasma electrons. Electron density perturbations, e.g. caused by the ponderomotive force $F_p = -\frac{q^2}{4m\omega^2} |\vec{E}(r,t)|^2$ of electromagnetic waves propagating in plasmas, induce electric fields antagonizing the separation. The electrons are accelerated in the potential and due to their inertia overshoot the regions of low electron density, leading to an oscillatory motion with frequency

$$\omega_{pe} = \left( \frac{n_e e^2}{\varepsilon_0 m_e} \right)^{1/2}$$  \hspace{1cm} (2.3)

where $n_e$ is the electron density, $e$ elementary charge, $\varepsilon_0$ vacuum permittivity and $m_e$ the electron mass. The plasma frequency can be mathematically derived from the continuity equation, Newton’s
second law and Poisson’s equation for electrostatics. In case of finite plasma temperatures, the oscillations can encroach to neighboring regions, leading to (longitudinal) electrostatic plasma waves. The dispersion relation of the electron plasma wave (also called Langmuir wave) is given by

$$\omega_{BG}^2 = \omega_{pe}^2 + \frac{3kbT_e}{m_e}k^2$$  \hspace{1cm} (2.4)

with $k_B$ the Boltzmann constant, $T_e$ electron temperature and $k$ wavenumber. Equation 2.4 is also known as Bohm-Gross dispersion relation. The second term is connected to the aforementioned condition that the plasma needs a finite temperature for an EPW to develop since then the group velocity $v_g = \frac{\partial \omega}{\partial k} \neq 0$.

Let us now consider ion acoustic waves in plasmas. The ponderomotive force equally acts on the plasma ions, but since $F_P \sim m^{-1}$ this force is reduced by at least three orders of magnitude. Therefore, ions rather respond to the electrostatic field of the induced electron distribution. The frequency of the oscillatory motion of the ions is

$$\omega_i = \left(\frac{n_i z^2 e^2}{\varepsilon_0 m_i}\right)^{1/2}$$ \hspace{1cm} (2.5)

with $n_i$ ion density, $z$ charge of the ion and $m_i$ ion mass. Comparing Eq. 2.5 with the frequency of the plasma electrons shows that the ion frequency is reduced by a factor of $\left(\frac{m_i n_i}{m_e n_e}\right)^{1/2}$. For typical plasma densities in this work e.g. $\frac{n_i}{n_e} \approx 0.03$. The dispersion relation of the ion acoustic wave under the assumption of negligible ion temperatures is simply

$$\omega^2 = c_s^2 k^2$$ \hspace{1cm} (2.6)

with $c_s = \left(\frac{Zk_B T_e}{m_i}\right)^{1/2}$ ion sound speed.

Having discussed the properties of particle waves, let us take a look at the effect of dispersion in plasmas on the propagation of electromagnetic waves. The ponderomotive force results in a motion of the electrons away from regions of high E-fields. EM waves traversing plasmas equally lead to
an oscillatory motion of the electrons in the direction of the E-field (laser polarization). Neglecting the magnetic field component the quiver velocity of the electrons is given by

$$v_{osc} = \frac{eE_0}{m_\epsilon\omega_0}$$

(2.7)

Note that the laser intensity can be expressed in terms of the electric field (neglecting influence of the B-field) by

$$I_0 = \frac{1}{2} c_\epsilon e_0 E_0^2.$$ Accelerated charges also radiate at the laser frequency but inherit a phase shift with respect to the driving frequency. This leads to the dispersion of EM waves in plasmas described by

$$\omega_L^2 = \omega_{pe}^2 + (ck)^2.$$  

(2.8)

Equation 2.8 implies that a light wave cannot propagate in a plasma if \( \omega_L < \omega_{pe} \). As a result, the critical plasma density can be defined as

$$n_c = \frac{e_0 m_\epsilon \omega_L^2}{c^2}.$$  

(2.9)

If plasmas are overcritical incident EM waves get reflected at the surface of the plasma. Especially in theoretical publications about plasma-based parametric amplifiers it is common to express the target density as the ratio between the plasma electron- and critical density \( N = \frac{n_e}{n_c} \).

The index of refraction for an EM wave is defined as the phase velocity divided by speed of light in vacuum \( n = \frac{c}{v_p} \). Using \( v_p = \frac{\omega}{k} \) and Eq 2.8 one can derive the refractive index of a plasma

$$n = \left(1 - \frac{\omega^2_{pe}}{\omega_L^2}\right)^{1/2} = \left(1 - \frac{n_e}{n_c}\right)^{1/2}.$$  

(2.10)

It follows that the refractive index of plasmas is exclusively < 1 and lower for increasing electron densities.
2.3 Parametric oscillators and instabilities

A parametric oscillator is a harmonic oscillator which is driven by a variation of one or more of its parameters, whereas the periodic variation has a different frequency compared to the resonant frequency $\omega_R$ of the oscillator. A classic example is a pendulum where the oscillation is driven by extending and shortening the arm of the bob with the driving frequency $\omega_D = 2\omega_R$. Initially small amplitudes of an oscillation can be amplified if driven by the correct frequency (parametric resonance).

Parametric instabilities occur for all materials (including plasma) when a periodic variation induces growing oscillations in a medium at a different frequency. This can only be the case if the interaction is nonlinear, e.g. for high power lasers.

In a plasma an intense laser pulse interacts (e.g. via the ponderomotive force) with the free charge carriers and can excite particle waves. Depending on the laser pulse duration the longitudinal plasma mode can be carried out by ions (ion acoustic waves) or electrons. Possible wave-wave interactions in a plasma are the decay instability ($n_e \approx n_c$), stimulated Brillouin scattering ($n_e < n_c$), two plasmon instability ($n_e = \frac{1}{4}n_c$) and stimulated Raman scattering ($n_e < \frac{1}{4}n_c$). The incident pulse can be backscattered from the self-induced particle wave (parametric backscattering instability).

2.3.1 Plasma-based optical parametric backscattering

The optical parametric processes of interest for the amplification of laser pulses are stimulated Raman backscattering (SRBS) and stimulated Brillouin backscattering (SBBS). They are three-wave instabilities, where the corresponding waves are the laser pump pulse, counterpropagating seed pulse and the plasma wave (Fig. 2.2). In case of SRBS the plasma wave relates to the electron plasma (alias Langmuir-) wave. In SBBS the pump and seed beam are coupled via an ion acoustic wave. The frequency and wavenumber matching conditions for both scattering processes are $\omega_0 = \omega_1 + \omega_{pl}$ and $k_0 = k_1 + k_{pl}$, where the indices 0, 1, pl denote the pump, seed and plasma frequency, respective wavenumber. The pump pulse must have a higher pulse energy and longer pulse duration than the seed pulse. The amplification process can be understood as follows, pump
and counterpropagating seed beam overlap in the plasma, the beat wave resonantly drives a plasma wave. The subsequent density modulation in the plasma acts as a phase grating leading to the Bragg reflection of the pump into the seed pulse. Since the duration of the amplified seed pulse can be much shorter than that of the pump pulse, orders of magnitude higher output powers are expected. Manley-Rowe relations dictate that the maximum energy transferred to the seed pulse is dictated by $1 - \frac{\omega_{pl}}{\omega_0}$ with $\omega_{pl}$ the frequency of the electron plasma-, respectively the ion acoustic wave. For typical plasma densities used in SRBS (SBBS) experiment this implies that 90 - 95% (> 99.9%) of the pump energy can be backscattered while the remaining energy is transferred to the corresponding plasma wave.

**2.4 Stimulated Raman backscattering**

Stimulated Raman backscattering (SRBS) in plasmas is based on the excitation of an electron plasma wave (EPW) by the beat of the pump and seed pulse. In order to resonantly drive the EPW the seed pulse requires a frequency shift $\omega_1 = \omega_0 - \omega_{pe}$. Since $\omega_{pe} \sim \sqrt{n_e}$ for a given central wavelength of the laser, the frequency shift is determined by the electron plasma density. However, the ratio between pump and EPW frequency $\frac{\omega_0}{\omega_{pe}}$ can not take arbitrary values if efficient driving of the SRBS instability is desired. In [41] PIC simulations define a parameter window of $14 < \frac{\omega_0}{\omega_{pe}} < 20$ where competing instabilities are minimized. Subsequent electron densities are

![Figure 2.2: Plasma-based parametric amplification scheme [41].](image-url)
10^{18} to the low 10^{19} cm^{-3} for Ti:sa (~800 nm) or ND:glass (~1050 nm) systems. The seed pulse then requires a shift of 40 – 80 nm to longer wavelengths usually realized by Raman crystals, solid-state optical parametric amplification or self-phase modulation (chapt. 3.3 and 5.2.1). In chapter 6 it is shown how high power seed pulses for future SRBS experiments can be generated.

The resonance condition for SRBS in terms of wavenumber of the plasma wave is [30]

\[ k_{pe} = 2k_0 \left[ 1 - \left( \frac{2\omega_{pe}}{\omega_0} \right)^{1/2} \right]. \tag{2.11} \]

If aforementioned resonance conditions are fulfilled, the growth rate of the SRBS instability in the linear case is

\[ \gamma_{SRBS} \approx \frac{1}{2} a_0 \left( \omega_0 \omega_{pe} \right)^{1/2} \tag{2.12} \]

with \( a_0 \) the vector potential of the pump. In the following chapter 2.4.2 \( \gamma_{SRBS} \) is compared to the growth rates of competing processes. The relation between the vector potential and intensity

\[ I_0 \left[ \frac{W}{cm^2} \right] = \frac{2.736 \times 10^{18} [a_0]^2}{\lambda^5 [\mu m]} \tag{2.13} \]

is used in this form for all upcoming calculations.

SRBS is a three-wave instability where the wave envelopes can be expressed by the following set of equations [24]

\[ \frac{\partial a_2}{\partial t} + c \frac{\partial a_2}{\partial x} = -\sqrt{\omega \omega_p} a_1 f \tag{2.14a} \]

\[ \frac{\partial a_1}{\partial t} - c \frac{\partial a_1}{\partial x} = \sqrt{\omega \omega_p} a_0 f^* \tag{2.14b} \]

\[ \frac{\partial f}{\partial t} - i\delta \omega f = \sqrt{\omega_0 \omega_p} \frac{a_0 a_1^*}{2} \tag{2.14c} \]

Here \( \alpha \) is the vector potential, \( f \) is the envelope of the EPWs electrostatic field and \( \delta \omega \) the detuning frequency (see chapt. 2.4.2). Equation 2.14b suggests that seed amplification can be increased by using higher pump intensities which holds true only before competing instabilities or other deleterious effects diminish the SRBS efficiency. PIC simulations in [41] identify pump intensities of \( \sim 10^{15} \text{ Wcm}^{-2} \) as an optimum value. However, best experimental results show an efficiency of
2.4 Stimulated Raman backscattering

~4% [94, 95] which is significantly lower than the 90 – 95% deduced from the Manley-Rowe relations or the tens of percent given by various simulations. Continuous experimental and theoretical studies are indispensable for a better understanding of the underlying processes limiting efficiency. Theoretical works on SRBS suggest numerous deleterious mechanisms that are discussed in chapter 2.4.2 and 2.4.3.

2.4.1 Amplification regimes

Various regimes of stimulated Raman backscattering, e.g. exhibiting different temporal or spectral characteristics, can be distinguished. In the linear regime the seed amplitude grows exponentially ($\sim e^{\gamma t}$) and is temporally stretched since the pulse maximum propagates with $c/2$ while the front of the pulse propagates with the speed of light [31]. The pump beam envelope $a_0$ is approximated to be constant during the interaction.

For high seed amplitudes the process enters the nonlinear regime where seed pulse energy grows linearly ($\sim \gamma t$) and temporal compression ($\sim \frac{1}{t}$) is achieved [110]. Temporal compression takes place since the seed depletes the pump pulse „shadowing“ its later parts from amplification. As a result, the seed maximum propagates with a superluminous velocity towards the leading edge. For strong pump depletion the seed pulse may transfer energy back to the pump which is then again able to amplify trailing parts of the seed and so forth. This can lead to a train of pulses with the first spike carrying the majority of the energy [31].

Another amplification regime, called superradiant Raman amplification, is accessed for sufficiently intense pulses ($4\omega_0^2 a_0 a_1 \geq \omega_{pe}^2$). Here the electron movement inside the plasma is determined by the ponderomotive force of the beat wave [11]. This regime is not as prone to resonance detuning and due to bandwidth broadening during amplification supports shorter output pulse durations ($\tau_1 \approx \frac{\pi}{4\omega_0^2 a_0 a_1}$) than linear stimulated Raman scattering [35].

A new regime was recently proposed by Farmer et al. where high seed intensities and pulse durations comparable to a plasma period ($\tau_1 \approx \frac{1}{\omega_P}$) lead to significant amplification even if the wavebreaking threshold is surpassed [55]. In the so-called coherent wavebreaking (CWB) regime of Raman amplification the peak of the short seed pulse is not affected by the phase-mixing that occurs with the onset of wavebreaking.
2.4.2 Competing instabilities, growth rates and detuning

Deleterious instabilities include Raman forward scattering (RFS) and Raman backscattering (RBS) of the pump before seed pulse interaction or high energy seed RFS. The growth rate for Raman forward scattering is

\[ \gamma_{\text{RFS}}(0,1) \approx \frac{1}{2\sqrt{2}} \omega_0 \alpha_0,1 \]  

(2.15)

while the growth rate for the stimulated backscattering instability is given by [30]

\[ \gamma_{\text{SRBS}} \approx \frac{1}{2} \sqrt{\omega_0 \omega_p \alpha_0}. \]  

(2.16)

Calculating growth rates for typical laser and plasma parameters in this work (\( I_0 = 10^{15} \) W cm\(^{-2} \), \( \lambda = 800 \) nm, \( n_e = 10^{19} \) cm\(^{-3} \)) yields \( \gamma_{\text{RFS}} \approx 7 \cdot 10^{10} \) s\(^{-1} \) and \( \gamma_{\text{SRBS}} \approx 5 \cdot 10^{12} \) s\(^{-1} \). RFS has about a two orders of magnitude lower growth rate but since the scattered wave travels in pump direction the instability has more time to develop. However, multidimensional PIC simulations show that for pump intensities < \( 10^{16} \) W cm\(^{-2} \) RFS is sufficiently small to be neglected [41].

The aforementioned instabilities can be mitigated by resonance detuning \( \delta \omega \), for example by large pump chirps or plasma gradients [29]. The detuning describes the deviation from perfect resonance of the waves \( \omega_0 - \omega_1 - \omega_p \neq 0 = \delta \omega \). It can be characterized by the dimensionless parameter

\[ q = \frac{\left( \frac{\partial \omega_p}{\partial x} - 2 \frac{\partial \omega}{\partial x} \right) c}{a_0^2 \omega_0 \omega_p}. \]  

(2.17)

describing the detuning growth rate of the pulse propagating in the backscatter direction [24]. \( \frac{\partial \omega_p}{\partial x} \) is especially high for chirped pulses originating from broadband lasers (e.g. Ti:sapphire systems).

Several theoretical publications suggest the usage of tailored plasma gradients to compensate pump chirp and subsequently exhibit lower detuning growth rates. The effect of plasma gradients and detuning on the amplification process is evaluated in chapter 5.
2.4 Stimulated Raman backscattering

Efficiency can also be lowered by optical nonlinearities such as self-focusing, self-phase modulation, resp. modulational instabilities in plasmas [25, 26].

Modulational instabilities are caused by relativistic dynamics causing a nonlinear refractive index. This can be the case if intense laser pulses accelerate the plasma electrons close to the speed of light. As a result of the introduced nonlinear refractive index self-focusing and self-phase modulation occur, which lead to deteriorating beam profiles, frequency detuning and subsequently lower efficiencies. For the amplified seed pulse the growth rate for the modulational instability is [63]

\[ \gamma_m = \frac{1}{2} \frac{\omega_p^2}{\omega_1} a_1^2 \tag{2.18} \]

which yields \( \gamma_m \approx 10^9 - 10^{10} \text{ s}^{-1} \) for the different amplified seed pulse parameters in this work. Since \( \gamma_m \ll \gamma_{SRBS} \) the effect is expected to be negligible.

Equally non-relativistic effects can cause self-focusing by generating a transversal density gradient via the ponderomotive force. The growth rate for subsequent filamentation of the pump, in the limit used in [98], is

\[ \gamma_{pf} = \frac{5}{4} a_0 \omega_{pi} \tag{2.19} \]

Which gives \( \gamma_{pf} \approx 10^{10} - 10^{11} \text{ s}^{-1} \) for typical values in the Jeti40 beamtime and is therefore more than one order of magnitude lower compared to \( \gamma_{SRBS} \).

It should be noted that some of the proposed restrictions to plasma-based parametric amplification in general (including SBBS, chapt. 2.5) are derived from theoretical calculations or simulations discussing the viability of those processes in context of next generation laser systems. Therefore they may not be significant in present day experiments where conversion efficiencies are too low to reach appropriate seed intensities.
2.4.3 Wavebreaking and Landau damping

Deleterious effects on the SRBS instability concerning the EPW are wavebreaking [27] and Landau damping [28]. Wavebreaking occurs if the quiver velocity of the electrons approaches the phase velocity of the EPW and implies heavy particle trapping by the wave potential. In [31] the wavebreaking threshold in terms of the vector potential is defined as

$$a_{wb} \approx \frac{1}{4} \left( \frac{\omega_{pe}}{\omega_0} \right)^{3/2}.$$  

With aforementioned parameters one calculates $a_{wb} \approx 0.0051$, which corresponds to an intensity threshold for the pump of $I_0 \approx 1.1 \cdot 10^{14}$ Wcm$^{-2}$. Typical pump intensities in the performed experiments are $\sim 10^{15}$ Wcm$^{-2}$. Wavebreaking is expected to have a negative impact on the SRBS efficiency for seed pulse durations ($\tau_1 > \frac{1}{\omega_{pe}}$) since then the coherent wavebreaking regime is not accessed. PIC simulations suggest that plasma densities near the wave breaking limit yield ideal gain in the nonlinear regime while output pulses maintain a viable temporal structure [34] since the leading spike of the backscattered pulse train is efficiently amplified instead of secondary oscillations. More recent simulations identify two different wavebreaking regimes [32]. A mild wavebreaking regime for pump intensities few times higher than the wavebreaking threshold where the efficiency drops to $\sim 30\%$ and a strong wavebreaking regime (intensity at least one order of magnitude above threshold) where efficiency can be reduced below $\sim 10\%$. For the latter case increasing seed intensities do not lead to higher SRBS efficiencies. After [32] all the results shown in this work are in the mild or strong wavebreaking regime.

Landau damping is a collisionless energy extraction from the EPW by the plasma electrons. Resonant electrons with a lower velocity than phase velocity of the EPW get accelerated while faster electrons get decelerated. Assuming a Maxwellian distribution, the quantity of lower velocity electrons is higher, thus more energy is extracted than gained. The electrostatic field amplitude of the EPW is lowered, reducing efficiency of the SRBS process (Eq. 2.14b). PIC simulations show that for tenuous plasmas ($n_e \leq 10^{19}$ cm$^{-3}$) Landau damping has no significant impact since it quickly runs into saturation due to a local flattening of the electron velocity distribution [41]. However, for high plasma temperatures ($T_e \geq 100$ eV) and dense plasmas ($n_e \geq 10^{19}$ cm$^{-3}$) Landau damping can affect Raman growth rates [37].
2.5 Stimulated Brillouin backscattering

As laid out in the paragraphs above, competing instabilities and deleterious plasma effects can be pronounced or mitigated by adjusting pulse or plasma parameters. A trade-off between increasing SRBS growth (achieved by high pump intensities, dense plasmas, small detuning) and restraining processes that diminish SRBS growth is required in order to extract amplified pulses with viable spatial and temporal structures at high efficiencies.

2.5 Stimulated Brillouin backscattering

Stimulated Brillouin backscattering (SBBS) in plasmas is based on the excitation of an ion acoustic wave (IAW) by the beat of the pump and seed pulse. The condition for frequency matching is \( \omega_1 = \omega_0 - \omega_{pi} \). Since the frequency of the IAW is comparably low (see chapt. 2.2) \( \frac{\omega_{pi}}{\omega_0} \leq 0.02 \) the seed frequency can be \( \omega_1 \approx \omega_0 \). Which means that at broadband (Ti:sa) the seed does not require a frequency shift.

The SBBS instability generally favors higher plasma densities compared to stimulated Raman backscattering. Typical densities are \( 10^{19} - 10^{20} \text{ cm}^{-3} \), whereas it is proposed to use \( n_p > \frac{1}{4} n_c \) in order to fully suppress competing Raman scattering instabilities [93]. However, other publications suggest using lower densities due to the onset of filamentation (see chapt. 2.4.2) at high intensities.

The resonance condition for SBBS in terms of wavenumber of the plasma wave is [30]

\[
N_G = 2N_a \left( 1 - \frac{\omega_{pi}}{\omega_0} \right) + \frac{1}{\omega_{pi}}. \quad (2.21)
\]

with \( k_0 = \frac{\omega_0}{c} \left( 1 - \frac{n_p}{n_c} \right)^\frac{1}{2} \) the wavenumber of the pump wave. The growth rate of SBBS can then be expressed as

\[
\gamma_{SBBS} = \frac{1}{2\sqrt{2}} \sqrt{\frac{c}{c_s}} a_0 \omega_{pi} \quad (2.22)
\]

where \( a_0 \) the vector potential of the pump, \( \omega_{pi} \) the frequency of the ion acoustic wave and \( c_s \) ion sound seed.
SBBS has long been disregarded as a viable amplification mechanism since the seed pulse duration is limited to ion acoustic timescales that are in plasmas around the picosecond range. However, pulse durations can be significantly shorter for SBBS in the so called strong coupling regime first described in a theoretical publication in 2006 [91].

2.5.1 Strong coupling regime

Strongly coupled SBBS (sc-SBBS) is described by a quasi-mode that is forced on the ion wave. It is therefore comparable to the superradiant Raman amplification (chapt. 2.4.1) where an electron plasma wave is equally not an eigenmode but a driven oscillation. In [91] the transition to the strongly coupled regime is defined by

\[
\left( \frac{v_{osc}}{v_e} \right)^2 > \frac{4k_0c_\omega \omega_0}{\omega^2_{pe}}
\]  

(2.23)

where \(v_e = \left( \frac{k_B T_e}{m_e} \right)^{1/2}\) the thermal velocity of the electrons and the oscillation velocity is here given in terms of the vector potential as \(v_{osc} = a_0 c\). If the condition is not fulfilled SBBS is in the weakly coupled regime and minimum output pulse durations are limited to \(\sim \frac{\lambda_0}{c_s}\). Sc-SBBS is favored for low plasma temperatures (few hundreds of eV) and high intensities (contributing to the electron quiver velocity). Once condition 2.23 is fulfilled the process becomes independent of temperature. In [37] equations for the strong coupling regime are derived under the stricter condition (compared to Eq. 2.23)

\[
\Lambda = \frac{\omega_{pe}^2 v_{osc}^2}{16k_0c_\omega \omega_0^2} \gg 1
\]  

(2.24)

where the ion plasma response is then given as

\[
\omega_{sc} = \left( \frac{1}{2} + \frac{i \sqrt{3}}{2} \left( \frac{k_0^2 v_{osc}^2 c^2 \omega_0^2}{2 \omega_0} \right) \right)^{1/3}.
\]  

(2.25)
The real part of $\omega_{sc}$ represents the frequency of the quasi ion wave which is typically slightly higher than $\omega_{pi}$. The imaginary part of $\omega_{sc}$ represents the (linear) growth rate $\gamma_{sc}$, whereas the compression of seed pulses down to $\frac{1}{\gamma_{sc}}$ is theoretically possible. This corresponds to shortest pulse durations in the two-digit femtosecond range.

Sc-SBBS is not as sensitive to density fluctuations as SRBS because of the low frequency of the ion wave (even for strong density fluctuations a broadband pulse will always find matching conditions) and the relaxed restrictions on frequency detuning. Figure 2.3 shows the frequencies of the EPW (black) and quasi ion mode (red) and clearly illustrates why density fluctuations lead to a much smaller detuning when the three-wave process couples to an ion wave. The robustness to fluctuations provides a better upscalability when changing to large focus diameters of the overlapping pulses which is inevitable if plasma-based parametric amplifiers should yield higher output powers compared to current CPA based laser systems.

Undamped linear growth rates for both instabilities are shown in Figure 2.4. From linear theory it follows that in sub-quarter-critical plasmas SRBS has much higher growth rates compared to sc-SBBS. However, the EPW is affected by Landau damping and wavebreaking prior to the ion wave (assuming $T_e \gg T_i$) and additional frequency detuning can significantly reduce the growth of the SRBS instability. Equally to SRBS, experimental results for sc-SBBS [40, 93] show conversion efficiencies of a few percent. Possible mechanisms limiting amplification are discussed in the following chapter.

![Figure 2.3: Frequencies of the electron/quasi-ion mode for $I_0 = 1 \cdot 10^{16} \text{ Wcm}^{-2}$ ($\alpha_0 \approx 0.048$), $\lambda_0 = 800 \text{ nm}$ and quasi-neutral $\text{H}_2^2+$ plasma.](image)
2.5.2 Deleterious effects in the strongly coupled regime

Assuming below quarter-critical densities $n_e < \frac{1}{4} n_c$ the Brillouin instability has to compete with the Raman instabilities (Eq. 2.15, 2.16). Though higher plasma temperatures in sc-SBBS experiments are expected to reduce Raman growth rates via Landau damping, it has been suggested that Raman forward scattering of the seed pulse may still be an issue [33]. [37] defines the ratio of seed to pump where RFS has to be considered

$$\frac{a_s}{a_0} = 1.94 \left( \frac{z_{me}}{a_0 n_i} \right)^{1/3} \left( \frac{\omega_0}{\omega_{pe}} \right)^{4/3}. \quad (2.26)$$

For this ratio the growth rates of RFS and SBBS are equal, it gives an estimate for the maximum seed intensity that can be obtained before the seed amplification is affected by detuning and
2.5 Stimulated Brillouin backscattering

envelope modulations. Subsequent PIC simulations have shown that for equal growth rates the seed amplification by sc-SBBS starts to saturate.

All Raman instabilities can be fully suppressed at plasma densities $n_e \geq \frac{1}{4} n_c$. However, new difficulties arise due to increasing growth rates of the filamentational instabilities (Eq. 2.18, 2.19). Therefore some theoretical publications only discuss amplification in sub-quarter-critical plasma densities [96]. However, in [33] it is proposed to use either $\frac{n_e}{n_c} > 0.25$ to suppress Raman instabilities (especially unseeded pump backscattering and seed RFS) or $\frac{n_e}{n_c} \leq 0.01$ where filamentation and Raman growth rates are low. The results for sc-SBBS in chapter 4 cover the latter condition with densities $\frac{n_e}{n_c} \approx 0.001 - 0.01$.

As in case of electron plasma waves (chapt. 2.2), ion plasma waves can equally experience (ion) particle trapping and subsequent wavebreaking. The growth rate for wavebreaking in the sc-SBBS regime can be expressed as

$$\gamma_{wb} = 2k_0 v_{osc} \left( \frac{m_e}{2m_i} \right)^{1/2}. \quad (2.27)$$

Wavebreaking reduces efficiency and can induce fracturing of the seed. It can be prevented for

![Figure 2.5: Threshold conditions for EPW wavebreaking, sc-SBBS and SRA regime depending on the electron plasma density and pump vector potential. Corresponding regimes are entered above the respective lines. Here $\lambda_0 = 800 \text{ nm}$, seed intensity $1 \cdot 10^{15} \text{ Wcm}^{-2}$ ($\alpha_i \approx 0.153$) and quasi-neutral H$_2^{2+}$ plasma.](image)
seed pulse durations $\tau_1 < \tau_{wb} = \frac{1}{\gamma_{wb}}$. At the same time $\tau_1$ should not be shorter than the inverse growth rate of the sc-SBBS instability given by the imaginary part of Eq. 2.25 $\left(\tau_{sc} = \frac{1}{\text{Im}[\omega_{sc}]}\right)$.

The thresholds for wavebreaking of the electron plasma wave and for entering the strongly couple stimulated Brillouin backscattering regime, respectively the superradiant Raman amplification regime are shown in Figure 2.5. For the calculation typical pulse and plasma parameters from the experiment introduced in chapter 4 are used.
Chapter 3

Seed pulse generation by nonlinear frequency conversion in Bessel beam pumped gas cells

This chapter introduces methods to exploit nonlinear optical effects in the context of seed beam generation for stimulated Raman backscatter experiments. Therefore the focus is on the generation of spectral components with lower frequencies. Ideally the central wavelength of the entire spectrum is redshifted at high efficiencies to the desired wavelength window required for phase-matching in SRBS.

Nonlinear optics is characterized by nonlinear responses of the dielectric polarization $\mathcal{P}$ in a material to the field of the electromagnetic wave. In general, when light propagates in a transparent medium, the field of the electromagnetic wave causes a copropagating electric and magnetic polarization wave inside the medium. Because of the much higher electric susceptibility, the magnetic polarization is usually neglected. The polarization is then often described by a power series expansion of the illuminating electrical field

$$\mathcal{P} = \varepsilon_0 \chi^{(1)} \mathcal{E} + \varepsilon_0 \chi^{(2)} \mathcal{E}^2 + \varepsilon_0 \chi^{(3)} \mathcal{E}^3 + \ldots$$  \hspace{1cm} (3.1)
with $\varepsilon_0$ the vacuum permittivity and $\chi^{(n)}$ the $n^{th}$-order susceptibility. For small electrical fields, respectively low intensities, the response of the medium is proportional to the field strength of the irradiating wave (first term in Eq. 3.1) and solely leads to a reduction of the phase velocity compared to vacuum propagation. This is described by the frequency dependent, dimensionless (linear) refractive index of the medium. For high field strengths, nonlinearities in the polarization occur and can lead to the generation of frequency components that are not present in the initial beam. As it is the case for the stimulated Raman scattering (SRS) process which can be described by a third-order nonlinear susceptibility $\chi^{(3)}$. SRS is a promising method to obtain redshifted pulses with high output powers. The usage of quasi-Bessel beams facilitates those high output powers due to increased gain lengths by orders of magnitude.

The chapter is finalized by a description of the temporal Kerr effect which equally arises from a third-order nonlinear susceptibility $\chi^{(3)}$. The temporal Kerr effect is related to the effect of self-phase modulation (SPM). In SPM a laser pulse can obtain new frequency components by the self-induced change of the intensity dependent nonlinear refractive index. Under certain conditions this can also lead to a shift of the central frequency.

### 3.1 Spontaneous Raman scattering

Spontaneous Raman scattering is an inelastic scattering process first discovered in 1928 by C.V. Raman. Illuminating a Raman active material leads to scattering of the incoming photons into lower (higher) frequency photons, called Stokes (anti-Stokes) radiation. In the context of this work Raman scattering in gases, thus inelastic scattering at free molecules is of interest. Raman scattering can equally appear in solids or liquids where the mediator of the energy transfer are optical phonons. In the case of a gas energy is transferred by the individual motions of the molecules.

For a molecule to be Raman active it must have an anisotropic polarizability (Gross selection rule). The energy difference of the incoming and scattered photon $\omega_{S/\text{aS}} = \omega_{L} \pm \omega_{ff'}$ is converted by means of a virtual intermediate level into an excitation or de-excitation of the molecule in terms of vibrational and or rotational modes (Fig. 3.1). $\omega_{L}$ denotes the frequency of the incident light. $\omega_{ff'}$ is the frequency difference between the vibrational and or rotational state of the molecule before and after the scattering process. $\omega_{S}$ and $\omega_{\text{aS}}$ are the frequencies of the scattered Stokes-, respectively
3.1 Spontaneous Raman scattering

Figure 3.1:
Schematic of the energy transitions for Stokes and anti-Stokes Raman scattering.

anti-Stokes radiation. The corresponding specific selection rules are $\Delta \nu = \pm 1$ for vibrational and $\Delta J = \pm 2$ for merely rotational transitions. The quantity of vibrational modes in a linear (nonlinear) molecule is $3N - 6$ ($3N - 5$) with $N$ being the number of atoms in the molecule. Generally, the most significant transition for Stokes radiation is a change from the vibrational ground mode $\nu_0$ to the energetically closest vibrational state $\nu_1$.

Frequency shifts are expressed in wavenumbers $\tilde{\nu}$ and reach from tens of cm$^{-1}$ for exclusively rotational to thousands of cm$^{-1}$ in case of vibrational transitions. Since frequency shifts of the scattered photons are characteristic for the molecule, Raman scattering has become of great importance in determining chemical compositions in the field of Raman spectroscopy. Stokes scattering is typically orders of magnitude more intense than anti-Stokes scattering because in thermal equilibrium the probability of finding a molecule in an excited state is reduced by $e^{-\frac{\hbar(\omega_f - \omega_g)}{kT}}$ (Boltzmann factor).

Spontaneous Raman scattering has a low cross section per unit volume, typically $1$ out of $10^6$ photons is Raman scattered while passing through $1$ cm of Raman active material. Not taking into account absorption and competing scattering processes, the spatial growth of the Stokes radiation for spontaneous Raman scattering is given by [66]

$$m_S(z) = m_S(0) + \frac{D m_L \eta_S z}{c}$$

(3.2)

where $m_S(0)$ is the photon occupation number for the Stokes mode at the entrance of the Raman media, $D$ material dependent proportionality constant, $m_L$ mean number of irradiating photons with angular frequency $\omega_L$, $\eta_S$ the refractive index at $\omega_S$ and $c$ speed of light. It follows that Stokes radiation grows linear with propagation length in the Raman active media.
Stimulated Raman scattering (SRS) was discovered shortly after the invention of the laser in 1962 due to a significant increase in the available intensity for the radiation of Raman active materials. SRS is a three-wave interaction process that couples two EM-waves to different energy levels in a medium. If the growth of the Stokes mode is sufficiently high or an additional wave with angular frequency \( \omega_S \) irradiates the Raman active media, the transition from spontaneous to stimulated Raman scattering (SRS) is made [67, 68, 69]. The threshold of SRS is usually defined as the point where 1% of the pump intensity is converted to the Stokes mode [87]. It is distinguished from the spontaneous process by emission into narrow cones in laser propagation or counter-propagation direction with an exponential growth and can be described by a third-order nonlinear susceptibility of the material.

The properties of SRS such as generating a directional, coherent output beam at high conversion efficiencies makes the process a feasible method for wavelength shifting of laser pulses. Common accessible wavelengths are restricted to the frequencies of laser active materials and the associated harmonics, respectively sum- and difference frequencies. SRS extended the attainable range significantly for continuous wave as well as short pulse laser.

Depending on the duration of the laser pulse \( \tau_L \) irradiating the Raman active media, different regimes can be accessed. In the transient regime, laser pulse durations are shorter than the dephasing time \( T_2 \) of the transition [70, 71, 72]. Dephasing describes the process when a system loses it coherence. In a classical picture one can imagine an ensemble of identical oscillators that are excited by an external force, through interaction with the ensemble individual oscillators may experience changes in phase or frequency which will lead to an overall decay of the correlation. If \( \tau_L \) is shorter or equal to the vibrational period \( T_1 \) of the transition, Raman interaction occurs in the impulsive regime (ISRS) [73, 74]. The different regimes exhibit different growth rates of the Stokes mode and or spectral characteristics.

For the remaining case, if \( \tau_L > T_2 \), the term steady-state SRS is applied. In the steady-state regime Stokes intensity can be approximated by [75]

\[
I_S(z) = I_S(0) \cdot e^{g_{ss}I_Lz} \tag{3.3}
\]

where \( I_L \) is the intensity of the pump laser (with angular frequency \( \omega_L \)) and \( g_{ss} \) the steady-state Raman gain coefficient.
The Raman gain coefficient can be expressed as

\[ g_{ss} = \frac{N \omega_S}{4 \Gamma n_S n_L m \omega_{ff} c^2 \varepsilon_0} \left( \frac{\partial a}{\partial Q} \right)^2 \]  

(3.4)

with \( N \) the total population density (upper and lower Raman transitions), \( \Gamma \) half-width at half-maximum of the Raman linewidth, \( n_S \) (\( n_L \)) refractive index of the Raman media for photons with angular frequency \( \omega_S \) (\( \omega_L \)), \( m \) effective reduced mass of the material oscillation, \( \omega_{ff} \) angular frequency of the Raman transition, \( \left( \frac{\partial a}{\partial Q} \right) \) hyperpolarizability for the Stokes wave and \( \varepsilon_0 \) the vacuum permittivity. In SRS, Stokes intensity grows exponentially with pump intensity \( I_L \) and interaction length of the pump and Stokes beam inside the Raman active material. Note that interaction length can be curtailed by a group velocity mismatch (GVM) induced temporal walk-off.

### 3.2.1 Transient stimulated Raman scattering

For pump pulse durations \( T_1 < \tau_L < T_2 \) Raman conversion occurs in the transient regime. Whereas typical dephasing times \( T_2 \) are in the range of picoseconds and in the case of Raman active gases dependent on molecule type and gas pressure. Dephasing times can be calculated from the measured linewidth of the Stokes transition by \( T_2 = (\pi \Delta \nu_{\text{FWHM}})^{-1} \). Typical values for vibrational periods \( T_1 \) are in the two-digit femtosecond range and solely depend on the type of vibrational transition. In the transient regime Raman gain is decreased and rather dependent on pulse energy than pump intensity [72]. The lower gain is due to the inertia of the molecular response, leading to a damping of the stimulated vibrations. Another result of short, intense pump pulse durations is the emergence of competing nonlinear optical effects due to high intensities, as self-phase modulation and self-focusing. Possible solutions to this issue are elongated foci reducing intensity while simultaneously increasing gain length.

For strongly transient SRS (\( \tau_L << T_2 \)) Stokes beam intensity at the output of the medium can be approximated by [76]

\[ I_S = I_S(0) \cdot \exp \left[ \frac{8g_{ss}}{\tau_2} \int_0^L \int_0^{\tau_L} I_L(z, t) \, dz \, dt \right]^{1/2} \]  

(3.5)
which shows that instantaneous pump intensity is not directly contributing to the Stokes gain, as in the case of the steady-state regime, but rather pump energy. Transient SRS has a tendency for pulse shortening, whereas the Stokes pulse is delayed with respect to the pump [59].

### 3.2.2 Bessel beam pumping

High pressure gas cells have long been used in various applications as a simple method to obtain Raman shifted pulses with comparably high intensities. Gases are preferable media for SRS in terms of keeping undesirable scattering losses and other nonlinear effects low which lead to reduced conversion efficiencies and deteriorating temporal and spatial beam profiles. Long optical interaction lengths (whilst exceeding SRS thresholds $\sim 10^9$ W cm$^{-2}$ [47]) are advantageous since peak intensities can be reduced and gain length increased at the same time. Spatially elongated focal regions can be generated by conical shaped lenses (axicons). Subsequent caustic lengths along the optical axis are prolonged by orders of magnitudes compared to the confocal parameters of Gaussian foci. Axicons, first proposed in 1954 [77], convert Gaussian beams into quasi-Bessel beams [78]. They gained popularity in recent years for their usage in microscopy [79], optical micromanipulation [80], material processing [81, 82], remote sensing [83] and ultrashort pulse generation [84].

Unlike Gaussian beams, Bessel beams are non-diffracting and therefore maintain an unchanged transversal distribution, which in good approximation holds true within the axicon's depth of focus (DOF) resp. caustic length. The caustic length (in terms of FWHM intensity distribution) can be approximated as

$$L = \frac{0.8 w_0}{(n_a-1)\tan \alpha}$$

(3.6)

where $w_0$ is the incident beam waist, $\alpha$ the axicon base angle and $n_a$ the refractive index of the axicon (Fig. 3.2). Behind the caustic separation of the k-vectors increases and on-axis intensity declines to zero leading to an annular (or doughnut-shaped) beam in the far field (Fig. 3.3).
3.2 Stimulated Raman scattering

The radial intensity distribution along the caustic for an initially collimated Gaussian beam can be approximated as [43]

\[
I(r, z) = 2\pi k (\tan^2 \alpha)(n - 1)^2 z I_0 \exp \left[ -2 \left( \frac{(n-1)z \tan \alpha}{w_0} \right)^2 \right] J_0^2 (k(n - 1)r \cdot \tan \alpha) \quad (3.7)
\]

where \( k \) is the wavenumber, \( I_0 \) incident on-axis intensity and \( J_0 \) Bessel function of the first kind (zeroth-order). Figure 3.4 depicts an example for the intensity distribution of a laser pulse in the DOF using Eq. 3.7.

Raman shifting by Bessel beams generates spatially separated Stokes and anti-Stokes, fundamental modes in the far field. While the annular fundamental and anti-Stokes modes propagate on a cone surface, the Stokes beam propagates on the optical axis and exhibits a slowly diverging Gaussian beam profile. This behavior can be described by a Cherenkov-type four-wave mixing process with the longitudinal phase
matching condition $2\vec{k}_0|| = \vec{k}_{S||} + \vec{k}_{as||}$ \[88\]. Here $\vec{k}_0$ is the wavevector of the fundamental laser radiation (perpendicular to the cone generated by the axicon), $\vec{k}_S$ the wavevector of the Raman scattered Stokes and $\vec{k}_{as}$ wavevector of the Raman scattered anti-Stokes mode (Fig. 3.5).

3.3 Self-phase modulation

The total refractive index of a material consists of a linear refractive index and a second term describing the instantaneous modification of the refractive index by an intense laser (known as Kerr effect)

$$n_{total} = n_0 + n_2 \cdot I$$

with $n_2 = \frac{3}{2n_0^2\epsilon_0c} \chi^{(3)}$ the nonlinear refractive index \[66\], and $I$ intensity of the laser pulse.

Spatially this results in self-focussing of pulses with a transverse intensity distribution $I(r)$ that decreases with radius (e.g. Gaussian pulses). The temporal intensity profile usually described by a gauss or sech\(^2\) distribution results in so called self-phase modulation (SPM). The time dependent phase delay can introduce a chirp on an initially unchirped pulse. It can equally modify the spectrum thus creating more frequency components commonly observed in fiber optics as spectral broadening of waveguided pulses. If the refractive index, as seen by the propagating pulse, has different absolute values for the leading and trailing edge a shift of the central frequency can be induced.
The nonlinear phase shift is proportional to the optical power and propagation distance in the media and given as

\[ \phi_{NL}(t) = \frac{2 \pi}{\lambda} n_2 \cdot I(t) \cdot L. \]  

(3.9)

Therefore long interaction lengths as can be realized in waveguides increase spectral broadening. Examples for waveguides are solid fibers, hollow-core fibers (HCF) or (expanded) plasma channels. The HCF guides light in the hollow region which is surrounded by a solid fiber material (cladding). HCFs are of particular interest due to high damage thresholds and the possibility to use various working gases at different pressures. SPM in a HCF was utilized for seed generation in the Jeti40 SRBS beamtime (chapt. 5).
Seed pulse generation by nonlinear frequency conversion in Bessel beam pumped gas cells
This section presents the results of the plasma-based parametric pulse amplification beamtime carried out at the Astra Gemini (Ti:sapphire) laser system (Rutherford Appleton Laboratories). As can be seen by Figure 1.3 in chapter 1, experimental data of parametric backscatter instabilities utilizing high pump energies and short seed pulses are rather rare. To the best knowledge of the author the set of pulse parameters in the Gemini beamtime is not covered by any other publication. Backscatter amplification experiments with multi-joule pump energies are not uncommon whereas concurrent short seed durations (~150 fs) and comparably high energies (~15 mJ) are. Besides that, all publications showing evidence of amplification in the strong coupling stimulated Brillouin backscattering (SC-SBBS) regime, which is suggested to be the main amplification mechanism in this beamtime, are performed at narrowband Nd:glass laser systems [40, 92, 93].

Figure 4.1 depicts the threshold conditions introduced in chapter 2 and shows were the experiment is located (red area). The majority of the data is acquired at high target densities and pump intensities (framed X). The threshold conditions for amplification in the sc-SBBS regime and EPW wavebreaking are clearly surpassed. Electron temperatures are assumed to be around 100 eV for the experimental parameters (pulse duration, intensity, plasma density) utilized in this campaign. For the most part the superradiant Raman regime is not expected to contribute to the amplification
process. It should be noted that contributions from stimulated Raman backscattering cannot be excluded, especially at low plasma densities and or pump intensities. Spectral gain characteristics are discussed in detail in section 4.3.4 and compared with simulations for different pulse and density parameters.

![Figure 4.1: Threshold conditions for different regimes. Red area represents pump pulse and target density parameters utilized during the beamtime. For the threshold calculations the laser wavelength $\lambda_0 = 800$ nm, seed intensity $1 \cdot 10^{15}$ Wcm⁻² ($a_1 \approx 0.0153$) and quasi-neutral H₂⁺ plasma are used.](image)

### 4.1 Astra Gemini laser system

Astra Gemini is a high power, ultrashort pulse laser system based on the CPA scheme. The front end consists of an ultrashort pulse oscillator providing pulses with an average power of 550 mW at a repetition rate of 75 MHz. The 12 fs output pulses are stretched to 7 ps in a glass block. In the kHz preamplifier the pulses are brought to the mJ-level. A fast Pockels cell scales down the repetition rate to 10 Hz. After the (transmission gratings) stretcher the 1 ns pulses are amplified to 1.2 J in three successive multipass Ti:sa crystals that are pumped by frequency
doubled ND:YAG lasers. Afterwards pulses are split in two and propagate through independent amplifier-compressor beamlines. The 90 x 25 mm Ti:sapphire amplifier rods are pumped by 60 J frequency doubled ND:YAG lasers. The small-signal gain per pass is around 4.2, after multiple passes each pulse has an energy of 25 J at a repetition rate of three shots per minute. The grating compressors have an efficiency around 60% and compress the top-hat, d = 150 mm, ∆λ = 30 nm pulses down to 30 fs. Output energy is 15 J and therefore peak power per pulse available on target is 330 TW.

The two beamlines give the possibility to adjust energy, spectrum and pulse duration independently for the two pulses. This makes Astra Gemini a suitable laser system to conduct plasma-based optical amplification experiments where pump and seed pulses with different parameters are required. Since the two pulses emerge from the same front end temporal jitter is not an issue for the temporal pulse overlap.

4.2 Setup and diagnostics

A CAD drawing of the setup is shown in Figure 4.2. It should be noted that the beampaths for the adaptive mirror-, shadowgraphy- and FROG diagnostic are not included in the Figure. The counterpropagating pump and seed pulse are in a 60 mrad off-axis geometry and therefore spatially separated for the different diagnostics.

The pump beam enters the meter scale target chamber on the top-left and is reflected by several dielectric mirrors until being focused by a f = 3 m, 12“ parabola. The converging beam is then sent to the target in a z-fold like geometry and traverses the focus in the 3 mm target region above the gas nozzle. After passing the target, the pump is reflected by several beamsplitters (1 x 3“ and 2 x 12“) to reduce the energy at the diagnostics. Outside the target chamber a 12“ concave dielectric spherical mirror focusses the diverging beam (Fig. 4.2 incorrectly suggests collimation). After transmitting another beamsplitter, the beam is focused onto an integrating sphere (ensuring spectral acquisition over the whole profile) connected to a an OceanOptics Maya2000 Pro fiber spectrometer.

The seed beam enters the target chamber on the top-right. It propagates through a 12“ linear polarizer, hits the adaptive optic and another 12“ mirror before being focused by a f = 3 m, 12“ parabola. After traversing the target region the seed pulse hits the 4“ beamsplitter. The reflection
hits a second 12” beamsplitter sending the beam to a concave $f = 3 \text{ m}$ dielectric spherical mirror for collimation. Outside the chamber a demagnifying telescope reduces the beam diameter appropriate to the 2” mirrors, beamsplitter and focusing optics set up on the respective diagnostic table. Latter couple the beam into the entrance slit of the Andor Shamrock303i spectrograph (coupled to an Andor Newton 920 CCD) and the 10x microscope objective ahead of the Andor iXon 888 CCD imaging the focal spot.

In the transmission direction of the 4” beamsplitter a 0.5” pick-up mirror extracts a part of the beam. This part is used for the pulse duration measurement by a SwampOptics FROG (8-20-USB). The pick-up mirror and additional propagation arm is required because the pulse has to exit the target chamber through a thin 1 mm window in order to keep the group velocity dispersion low.

The remaining (nearly full beam diameter) part of the seed exits the target chamber and hits the pyroelectric energy sensor that is imaged by an additional CCD.

The probe pulse for the shadowgraphy diagnostic is separated from the seed between the 12” mirror and seed focusing parabola (before entering the target chamber expansion). Again a 0.5” pick-up mirror is used to extract a part of the seed and send it to an additional propagation arm, consisting of multiple 1” mirrors, a delay stage and focusing optics in a 4f configuration. At last, the probe hits the CCD camera outside the target chamber that images the target plane perpendicular to the seed-pump propagation direction.

For the sake of completeness it should be mentioned that the pump backscatter arm (exiting target chamber top-middle) was abandoned since the cone angle of solitary pump backscattering is sufficiently wide to hit the 4” beamsplitter dedicated to the seed diagnostics.
4.2 Setup and diagnostics

Figure 4.2: CAD drawing of the experimental setup.
4.2.1 Seed and pump pulse parameters

As mentioned before, Astra Gemini has two independent amplifier-compressor beamlines. One beamline delivers the pulse that is used as a pump pulse and the second beamline delivers the seed pulse.

The pump pulse durations for this experiment are $\tau_0 = 6.5, 10, 13$ ps. It is possible to over-compensate the chirp from the stretcher to obtain negatively chirped pulses. A pulse duration of 6.5 ps with a negative chirp are set for one of the measurement series. For geometric reasons the pump pulse diameter is halved resulting in a beam diameter of 75 mm and energy up to $E_0 = 3.6$ J. The FWHM bandwidth is $\Delta \lambda = 30$ nm with a central wavelength $\lambda_c \approx 800$ nm (Fig. 4.3).

The second amplifier-compressor beamline delivers the seed beam. Spatial chirp in the seed compressor is utilized to cut the high frequency parts of the spectrum. As a result, redshifted seed pulses at central wavelengths between $\lambda_c = 806 – 812$ nm and bandwidths of $\Delta \lambda = 15 – 25$ nm (depending on the metal plate position between the compressor gratings) are obtained. Output pulse energies are up to $E_1 = 16$ mJ with a pulse duration $\tau_1 \approx 150$ fs.

Both beams are focused by 3 m parabolas. Corresponding f-numbers are $f/40$ and $f/20$, measured beam diameters in the interaction region are $w_0 \approx 70$ $\mu$m and $w_1 \approx 100$ $\mu$m for the seed after moving out of best focus in order to overspread the pump diameter. The adaptive mirror in the seed arm ensures a high-quality beam profile at the pulse overlap position. Profiles of both beams, exhibiting a nearly perfect Gaussian behavior, are shown in Figure 4.4. Peak pump intensities are $I_0 = 1.3 \cdot 10^{15} – 1.4 \cdot 10^{16}$ Wcm$^{-2}$, while seed intensities range from $3 \cdot 10^{14}$ to $1.3 \cdot 10^{15}$ Wcm$^{-2}$.

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Figure 4.3: Pump spectrum (green) and seed spectrum (red) after compressors, dashed lines indicate the central frequencies.

Figure 4.4: Focal spots of pump and seed beam at target position.
4.2 Setup and diagnostics

4.2.2 Spatial and temporal overlap

Plasma-based parametric optical amplifiers require spatial and temporal overlap of the two pulses at the target position.

The spatial overlap is established by imaging a fine crosswire above target center with two independent focus diagnostics. Each consisting of a 10x microscope objective and a vacuum compatible CCD camera mounted on a motorized linear stage. The beams are attenuated by a waveplate and one after another is put at the crosswire position by alternating the focus diagnostic.

Temporal overlap at the target with a two-digit picosecond accuracy is found in a first step by fast photodiode measurements. Since seed pulse intensity is sufficient to ionize the gas jet, a more precise overlap can be established by using the probe shadowgraphy diagnostic. Two delay stages are required for this procedure, one is located in the seed beamline adjusting overlap relative to the pump pulse. A second delay stage is located in the probe arm inside the target chamber to adjust the timings when the probe pulse images the density disturbances. Having a short probe pulse duration, this method narrows the delay error approximately to the low picosecond to 3-digit femtosecond range. The density disturbances caused by pump and seed beam separately are shown in Figure 4.6. After finding the proper delay setting, where the probe pulse sees the disturbances emerging at both pulse entrance sides, fine-tuning of the delay is carried out by amplification scans (chapt. 4.3.2).
4.2.3 Gas jet target

The target is a 3 mm conical nozzle mounted on a XYZ-translation stage. It generates the hydrogen gas jet inside the target chamber where the two pulses overlap. The valve of the nozzle is triggered 3 ms before pulse interaction (opening time 4 ms) to ensure laminar gas flow. The backing pressure at the valve ranges from a few to 28 bar. The backing pressure determines the neutral gas density and subsequently electron density inside the plasma. It is ensured that the leading edge of the high energy pump pulse is sufficient to fully ionize the hydrogen gas. For full ionization the resulting electron densities in 1 mm height above nozzle are expected to range between $n_e = 2 \cdot 10^{18} \text{ cm}^{-3} - 1.6 \cdot 10^{19} \text{ cm}^{-3}$ as follows from FLUENT simulations performed at Strathclyde university (Fig. 4.7).

![Figure 4.7: Simulations of the electron densities in different heights above the 3 mm orifice at a backing pressure of 12 bar (hydrogen gas, fully ionized).](image)

4.3 Experimental results

The energies of the amplified seed pulses $E_{\text{gain}}$ are given as the total energy $E_{\text{tot}}$ subtracted by $E_{\text{BS}}$ and $E_{1,\text{tr}}$. Where $E_{\text{BS}}$ is the pump backscattering energy measured for blocked seed pulses at the respective backing pressure $p$, pump energy $E_0$ and pump duration $\tau_0$. $E_{1,\text{tr}}$ is the seed energy transmitted through the target measured for blocked pump pulses, equally at the respective pressure, energy $E_1$ and duration $\tau_1$. Seed transmission does not significantly increase because of
4.3 Experimental results

plasma channel generated by the pump pulse which was verified by measurements of the transmitted energy for seed delays where the pump fully traversed the target before seed arrival. For a better understanding, Figure 4.8 shows the data for the different energies depending on the backing pressure at the gas nozzle. Plasma generation and subsequent scattering losses continuously decrease the seed transmission for higher pressures. At $p = 4.5$ bar energy transmission is already decreased to ~75% compared to the initial seed energy. The (unseeded) pump backscattering on the other hand increases for higher target densities, respectively plasma electron densities. The total energy $E_{\text{tot}}$ is measured for best amplification (optimized pulse overlap position inside the target). It includes the backscattering energy from the pump, seed energy transmitted through the plasma and the energy gain of the seed pulse induced by the stimulated backscattering instability in the following called $E_{\text{gain}}$.

Shot-to-shot fluctuations lead to standard deviations of the total energy output up to ±20% with maximum errors up to 50%. A direct correlation to the pump energy fluctuations (<10%) could not be found. In the majority of cases several shots showing distinct amplification are selected and averaged in order to show the amplified spectra or output energies.

The plotted seed spectra (acquired by the Andor spectrograph) are corrected in terms of background, CCD quantum efficiency and the utilized neutral density filters, as well as the optical components inside the spectrograph including two gold gratings (150 ln/mm or 600 ln/mm) and several aluminum mirrors (Fig. 4.9).

Figure 4.8: Pressure dependent energies measured by the pyroelectric energy sensor in the seed direction for an unseeded pump, seed transmission through target and total output energy for the amplification shots.
4.3.1 Spontaneous pump backscattering

Because of the high pump pulse energies a significant part is backscattered from the plasma without any seed interaction. The backscattering instability grows from noise in the plasma and is also referred to as spontaneous backscattering. Without the injection of a seed pulse, backscattering (BS) is directed opposed to the incoming pulse. Since the setup is in a 60 mrad off-axis geometry, the backscattering should not be picked up by the seed diagnostics. However, plasma effects can cause a change to the beam divergence, resulting in a widened backscattering cone (Fig. 4.11). An outer part hits the optics dedicated to the seed diagnostics and subsequently the measurement devices. This is demonstrated in Figure 4.10, where nearfield profiles of the seed and backscattered pump beam are shown. The black spots in the seed beam profile are due to the pick-up mirrors for the initial seed spectrum (left), probe beam (right) and pulse duration measurement (top). The sharp circular contour of the backscattered pump profile is caused by the 4° beamsplitter mount. Highest intensity is seen on the right-hand side since it is the side of the incoming pump pulse. Spontaneous backscattering spectra can be an indicator of the electron densities in the plasma. In the linear case the frequency resonance condition \( \omega_1 = \omega_0 - \omega_{pl} \) determines the frequency of the backscattering.
backscattered pulse. The frequency of the pump $\omega_0$ is known by default, $\omega_1$ is determined by the spectrograph measuring the BS wavelength ($\omega_1 = \frac{2\pi c}{\lambda_1}$). In case of a backscattered Raman signal the electron density can be calculated by $n_e = \frac{(\omega_0-\omega_1)^2\epsilon_0m_e}{e^2}$. The backscattering spectra show characteristics of EPW based parametric backscattering (Raman backscattering).

The development of the unseeded backscattering spectra of the pump pulse ($E_0 = 3.5$ J, $\tau_0 = 6.5$ ps) versus gas pressure are shown in Figure 4.12. Each spectra is averaged over three shots and the initial pump spectrum (normalized to the highest signal count) is plotted for comparison. Vertical dashed lines indicate the central frequency of the backscattering peaks. A calculation of the electron densities then yields $1.5 \cdot 10^{18}$ cm$^{-3}$ (10 bar), $2.3 \cdot 10^{18}$ cm$^{-3}$ (20 bar) and $3 \cdot 10^{18}$ cm$^{-3}$ (28 bar). The calculated densities differ greatly from the expected values given by the FLUENT simulations and experiences from previous experiments with similar nozzles. Also a linear dependency of the neutral gas density on the backing pressure is anticipated but not seen here. Possible explanations for the severely frequency shifted backscattering spectra are incomplete ionization, backscattering from the low density regions at the nozzle edges, the expulsion of electrons by ponderomotive forces, the Langmuir decay instability and particle trapping. The first argument can be ruled out since full ionization ($H_2^{2+}$) should be established in the leading edge of the pump pulse as PPT code calculations for similar parameters suggest [99].

In the Langmuir decay instability the EPW decays into a secondary EPW and an ion acoustic wave. The coupling between the two waves can lead to a reduction of the EPW frequency. This effect can only account for wavelength shifts of a few nanometers and is not expected to be the main shifting mechanism here [101].

Further possibilities are the reduction of the plasma density by the traversing pump beam expelling electrons via ponderomotive force from high intensity regions so that later parts of the pump experience smaller frequency shifts. Pump timescales are rather short so that the expulsion of electrons by the ponderomotive force should be negligible.
Particle trapping applies when $k_{pe}\lambda_D \geq 0.29$, with $\lambda_D = \left(\frac{e_0 k_B T_e}{n_e e^2}\right)^{\frac{1}{2}}$ the plasma Debye length for negligible ion temperatures [102]. The condition is fulfilled in the low density case (10 bar backing pressure) for electron temperatures $T_e \geq 40$ eV and at highest pressures ($p = 28$ bar) for $T_e \geq 110$ eV. Such temperatures are accessed by inverse Bremsstrahlung absorption of the pump pulse. At initially small electron temperatures the absorption rate is around $0.2$ eV/fs at $I_0 = 1 \cdot 10^{16}$ W/cm$^2$ which corresponds to the intensity of the pump beam in Figure 4.12 [103]. Though the value calculated by aforementioned publication is given for a few times higher plasma densities, electron temperatures of $T_e \geq 110$ eV are likely to be reached. Therefore, particle trapping could indeed contribute to the frequency shift in the spectra. However, it cannot account for the shifting of tens of nanometers [102].

An additional solid angle dependence of the backscattering spectra cannot be ruled out. Wavefront bowing of the EPW can lead to widened backscattering cones [104]. It was shown that the nonlinear frequency shift with concurrent wavefront bowing (induced by particle trapping) generates an angle-dependent shift of the backscattered spectra [109]. The simulations in aforementioned publication were made for the strong kinetic regime $k_{pe}\lambda_D \geq 0.35$ at much higher plasma densities and electron temperatures and show frequency shifts that are equally insufficient to account for the deviations of up to 60 nm obtained here.

[100] shows saturation of the RBS wavelength for increasing pressures in a 2 mm flat-top density gas jet and suggest early critical self-focusing at the low density boundary of the gas jet followed.
by a termination of the Raman backscattering before the high density plateau is reached. Pump pulse powers for the measurements in Figure 4.12 do not surpass the critical power for self-focusing \( P_c \,[\,GW\,] = 17.4 \cdot \left( \frac{\delta_n}{\alpha_p} \right)^2 \). Since the pump beam is focussed in an f/40 geometry the Rayleigh length \( z_R = \frac{\pi w_0}{\lambda} \) is around 3 mm which is comparable to the target length so that no critical self-focussing is required to obtain high intensities at the boundary of the gas jet. A termination of the Raman backscattering after the density ramp as proposed in [100] is not expected since intensities are not sufficient for cavitation and bubble formation wavebreaking occurring in laser wakefield acceleration experiments.

A more simple and straightforward explanation for the unexpectedly high frequency backscattering are actual low target densities which can be caused by decrepit valves. Fatigue phenomena of the valve can delay timeframes for complete opening and effect the formation of the density profile. Also, an incomplete valve opening can reduce the target densities especially at high backing pressures. However, a faulty valve is not expected to explain the high frequency backscattering spectra since gas nozzle / valve are comparably new, the triggering of the valve 3 ms before pulse arrival should compensate for small delays of the valve opening process and it is actually possible to measure spectral components in the backscattering spectra representing the expected target densities from the FLUENT simulations as will be shown in the following.

The spectra in Figure 4.12 have extended broad shoulders on the low frequency side. The shoulder

![Figure 4.13: Extended pump backscattering spectra for different backing pressures. Black dashed lines show the positions where the spontaneous Raman peak of a \( \lambda = 800 \) nm pulse is expected for the target densities at corresponding backing pressures following from the FLUENT simulations.](image)
extends further for higher gas pressures and is more of an indicator of the actual electron densities reached in the target. To investigate the full extent of this shoulder the spectrograph grating is exchanged by a grating with greater line distance. This reduces spectral resolution but allows for a broader wavelength range to be acquired. The results are shown in Figure 4.13. The dashed lines indicate the backscattering wavelengths for the different pressures as given by the FLUENT simulations. Good agreement between experiment and simulation is found when considering the low frequency parts of the backscattered spectra. The shift to longer wavelengths compared to the simulations may be explained by the broad bandwidth of the pump and or longitudinal heating of the plasma electrons which increases the frequency of the EPW. A similar wavelength shift of the Raman peak is observed during the Jeti40 beamtime (chapt. 5).

4.3.2 Energy gain and spectral dependence on relative pulse delays

The temporal overlap at the target is established by the shadowgraphy diagnostic. To maximize seed amplification, a more precise adjustment of the relative pulse delay is required. The relative delay determines the overlap position of seed and pump pulse in the target. Figure 4.14 shows the normalized energy gain $E_{\text{gain}}$ at different relative pulse delays $t_d$ between pump and seed pulse. The pump parameters are $E_0 = 3.5$ J and $\tau_0 = 13$ ps. The initial seed pulse energy $E_1 = 13$ mJ is reduced to $\sim 8$ mJ after propagating through the (p = 10 bar) hydrogen target.

![Figure 4.14: Seed amplification depending on the overlap position / relative delay of seed and pump pulse ($\tau_0 = 13$ ps, $n_e \approx 6 \cdot 10^{18}$ cm$^{-3}$).](image)
without pump presence. Seed pulse duration is ~150 fs. The delay scan shows that high seed amplification is reached in a small delay window of around $\Delta t_d = 2$ ps which corresponds to ~1200 $\mu m$ additional propagation length introduced by the delay stage. A stepsize of 500 $\mu m$ or less is suggested in order to scan the overlap position.

The zero relative delay is arbitrary set to the point of best amplification. Positive (negative) $t_d$ values represent the case when the seed pulse arrives late (early) related to the overlap for best amplification. For $t_d \approx -5$ ps the seed exits the target when the leading edge of the pump arrives at the boundary. Due to a lack of pulse overlap in the plasma the seed experiences no amplification. For $t_d > -5$ ps pulse overlap in the plasma is established. Best amplification is found at $z \approx \frac{1}{2} z_t$, with $z_t$ target length. At this particular position the seed pulse is expected to interact with the leading edge of the 13 ps pump pulse. The extended shoulder for positive $t_d$ can be explained by a small increase in seed transmission since the effects of ionization scattering and inverse Bremsstrahlung on the seed amplitude should be reduced when traversing a plasma that is already heated by the pump pulse.

A schematic of the estimated pulse overlap for the case $t_d = 0$ is shown in Fig. 4.15. Grey shaded area represents the gas target region, seed pulse overlaps with the pump peak intensity in the target is evident. Though it follows from Figure 4.15 that pulse interaction (in terms of pump power seen by the seed) may be optimized by increasing the relative delay by a few picoseconds. However, frequency detuning seems to lead to an overall reduction of the energy gain. This is supported by pulse delay measurements for negative pump chirps where a double peak of the energy gain is exhibited (Fig. 4.16). Unfortunately it was not possible to directly compare the relative delays at different pump chirps because of the undefined change in path length when moving the compressor grating. Subsequently, the zero position is arbitrary set to the first energy gain peak. It is suggested that this peak corresponds to an overlap with optimized pump power interaction while the second peak (at ~3 ps) is likely due to optimized frequency matching for the sc-SBBS process which is suggested to be the dominant energy transfer mechanism.

![Figure 4.15: Schematic of the temporal pulse overlap in the target (shaded area) for the optimized energy gain at zero delay.](image-url)
Figure 4.16: Energy gain at different relative delays for a negatively chirped pump pulse ($\tau_0 = -6.5$ ps, $n_e \approx 6 \cdot 10^{18}$ cm$^{-3}$).

Figure 4.17 shows the corresponding backscattering spectra at three different relative delays $t_d$. The spectra are averaged over three shots and a Gaussian filter is applied to remove spectral spikes for better comparability. The red dashed line shows the initial seed spectrum (before target interaction). At -5 ps (blue curve) a broad high frequency shoulder appears. While traversing the target, the seed ionizes the gas. The seed pulse generates a positive electron density gradient $\frac{dn_e}{dt} > 0$ at its ionization front and as a result a negative refractive index gradient (Eq. 2.10) which induces spectral blueshifting via SPM [105]. At $t_d = 0$ ps (yellow curve) the seed enters the target and within a short distance starts to propagate in the plasma generated by the leading edge of the seed. The amplification spectra for different relative pulse delays $t_d$ directly correspond to the measurement shown in Fig. 4.14 ($\tau_0 = 13$ ps, $n_e \approx 6 \cdot 10^{18}$ cm$^{-3}$).
pump, hence the extended high frequency shoulder is lowered. This effect is even more pronounced if the seed traverses the target when the pump pulse already passed the target (green curve). All amplification spectra (for $-5 \text{ ps} < t_d < 13 \text{ ps}$) look similar in terms of the spectral peak intensity and seem to not depend on the relative pulse delay (not shown here). This is fundamentally different to the observations made in other stimulated Raman backscattering experiments where relative delay depended wavelength shifts are significant [63] (see also chap. 5.3.1).

### 4.3.3 Increasing energy gain and efficiency for high pump intensities and plasma densities

This section treats the seed energy gain for different pump pulse energies and chirps. Pump energy is controlled by the energy of the frequency doubled ND:YAG laser generating the population inversion in the Ti:sapphire amplifier crystal. Pump energies are varied between $E_0 = 0.6 - 3.5 \text{ J}$. Pump chirp is controlled by moving the compressor grating, subsequent durations are $\tau_0 = -6.5, 6.5, 10, 13 \text{ ps}$. The amplification is expected to increase with higher pump energies as long as no deleterious effects (e.g. wavebreaking, RFS, filamentation) start to diminish the backscattering instability. For long pump pulses ($\tau_0 = 13 \text{ ps}$) an exponential increase of the output energy is measured (Fig. 4.18). The energies are averaged over three shots and initial seed parameters are $E_1 = 13 \text{ mJ}$ and $\tau_1 = 150 \text{ fs}$. The curves suggest that no saturation of the

![Figure 4.18: Seed energy gain dependence on pump energy for $\tau_0 = 13 \text{ ps}$.
Measurement points averaged over three shots. Plasma densities are $6 \cdot 10^{19} \text{ cm}^{-3}$ (10 bar) and $1.2 \cdot 10^{19} \text{ cm}^{-3}$ (20 bar).](image-url)
amplification is reached, so that higher pump energies could in principle lead to higher output energies. Deleterious instabilities are not yet seen to affect the amplification process. Quite the contrary, exponential energy gain with $E_0$ proofs that efficiency is still increasing. For the shorter pump pulse duration $\tau_0 = 6.5$ ps seed energy gain grows exponentially until $E_0 \approx 1.6$ J from where it exhibits linear growth (Figure 4.19). Calculating the peak pump intensity at this point yields $I_0 \approx 6.5 \cdot 10^{15}$ Wcm$^{-2}$. The peak pump intensity from Figure 4.18 reaches $7 \cdot 10^{15}$ Wcm$^{-2}$, which may explain why no transition to linear energy growth is observed. PIC simulation in [41] show that Raman forward scattering and filamentation destroy the seed profile during amplification for pump intensities $\sim 10^{16}$ Wcm$^{-2}$. RFS of the seed or pump pulse is not observed in this beamtime (for all parameters). Equally filamentation of the seed pulse profile is not observed with the far-field diagnostic, even for highest initial pump $\sim 1.4 \cdot 10^{16}$ Wcm$^{-2}$ and highest seed output intensities. Therefore, those two processes are not considered as mitigating energy transfer and being responsible for a reduction in energy gain. A quick estimate shows that the linear increase seen in Figure 4.19 is solely due to higher pump energies and not an increase in efficiency of the amplification process. Energy transfer efficiency can be increased for intense ($\sim 10^{16}$ Wcm$^{-2}$) pump pulses when utilizing higher plasma densities (Fig. 4.20). Here, at a plasma density of $n_e \approx 1.6 \cdot 10^{19}$ cm$^{-3}$ (0.009$n_c$) an increase of the efficiency is still observed when increasing the pump intensity from $1 - 1.4 \cdot 10^{16}$ Wcm$^{-2}$. Figure 4.19: Seed energy gain dependence on pump energy for opposite chirp signs of a 6.5 ps pump pulse duration. Every measurement point is averaged over three shots. Plasma density $n_e \approx 6 \cdot 10^{18}$ cm$^{-3}$. Figure 4.20: Increase of energy gain and efficiency at high pump intensities and target densities ($\sim 1.6 \cdot 10^{19}$ cm$^{-3}$). Measurement points averaged over three shots.
4.3.4 Spectral gain characteristics and amplification regimes

The purpose of this section is to deliver additional arguments why the main amplification process is suggested to be strongly coupled stimulated Brillouin backscattering. Focus is laid on the spectral gain characteristics of the seed pulses.

As shown in chapter 2 (Fig. 2.4) Raman growth rates in sub-quarter critical densities exceed sc-SBBS growth rates. However, this assumption neglects deleterious effects on the EPW that are expected to reduce the efficiency for Raman backscattering. In [37] PIC simulations show that for target densities ~0.009n_c the Raman growth rate exponentially declines from $T_e \geq 60 \text{ eV}$ due to Landau damping. Electron temperatures surpass this value while the pump traverses and heats the plasma via inverse Bremsstrahlung.

The deleterious effect of EPW wavebreaking is discussed in chapter 2.4.3. The pump intensity is a multiple of the intensity threshold for wavebreaking, therefore strong wavebreaking accompanied by a significant reduction of the SRBS efficiency is expected [32]. In contrast to that the seed pulse is not prone to wavebreaking of the quasi ion mode (coupled to the sc-SBBS instability). The time constant for ion wavebreaking is $\tau_{wb} \approx 320 \text{ fs}$ (Eq. 2.27) for highest pump intensities. Since $\tau_1 < \tau_{wb}$ this should not negatively effect growth of the seed in the sc-SBBS regime.

Finally, the detuning within the frame of SRBS is severe, especially at high plasma densities. The shift of the central wavelength between seed and pump is only around ~10 nm (Fig. 4.3). For a backing pressure of 28 bar (~$1.6 \cdot 10^{19} \text{ cm}^{-3}$) optimized seed gain is expected at ~890 nm. As can be seen in the following, amplification is predominant where the spectral intensity of the seed is high ($805 – 815 \text{ nm}$). Even when taking the bandwidth of both pulses into account there should not be any spectral components remotely satisfying the SRBS frequency matching condition. This significantly diminishes SRBS efficiency since for the most part the threshold to the superradiant regime (allowing for a more broadband spectral gain) is not surpassed.

Sc-SBBS matching can be calculated by the real part of Eq. 2.25 and yields a wavelength shift of only a few nanometers ($\Delta \lambda \leq 3 \text{ nm}$) for the present experimental parameters. The proximity of seed and pump spectra ensure good frequency matching coupled to the quasi ion mode.

Figure 4.21 shows the backscattered spectrum of an unseeded 3.5 J, 6.5 ps pump (green curve), the transmitted seed spectrum in absence of the pump beam (red curve) and the spectrum measured for pulse amplification at an optimized relative pulse delay (blue curve). The electron density is $1.6 \cdot 10^{19} \text{ cm}^{-3}$ (~0.009n_c). All spectra are averaged over three shots. Transmitted seed energy is ~4 mJ
Figure 4.21: Seed transmission (red), spontaneous backscattering (green) and output spectrum for pulse overlap in the target (blue). Pump pulse energy is \( E_0 = 3.5 \text{ J} \) (\( \tau_0 = 6.5 \text{ ps} \)). Initial seed energy \( E_1 = 13 \text{ mJ} \) (\( \tau_1 = 150 \text{ fs} \)), after transmission \( E_{1,tr} = 4 \text{ mJ} \). Average total output energy for the amplified shots is 75 mJ where average spontaneous backscattering amounts to 18 mJ. Backing pressure is at 28 bar which results in an electron density \( 1.6 \cdot 10^{19} \text{ cm}^{-3} \).

which is \(~30\%\) of the initial seed energy. The majority of the unseeded backscattering spectra is found between 830 – 840 nm, the seed spectrum stretches from \(~795 – 820 \text{ nm}\). Total output spectrum for the pulse overlap (amplification) shots shows strong spectral gain where the initial spectral seed intensity is high.

At the same time the spectra indicate a small amplification in the wavelength range around the peak of the Raman backscattered spectrum (around 835 nm). The overall amplification could be a hybrid of SRBS and sc-SBBS, whereas the Brillouin process is much more efficient in terms of energy transfer. This is in agreement with PIC simulation showing that in case of sc-SBBS amplification SRBS is only relevant in the initial stage of the pulse overlap and its contribution to the seed output energy is significantly smaller [92].

We define the spectral gain similar to the energy gain (see chapt. 4.3). Spectra of amplified shots are subtracted by the (unseeded) backscattering and seed transmission spectra. Figures 4.22 – 4.24 show the development of the spectral gain for different plasma densities, pump energies and pump chirps. Lower backing pressures result in lower plasma densities whereas 4.5 bar correspond to a plasma density of \(~2.6 \cdot 10^{18} \text{ cm}^{-3} \) (0.0015\( n_c \)). At low plasma densities the amplification in the wavelength window corresponding to Raman backscattering (~820 – 850 nm) is relatively more significant and in the wavelength window of high spectral seed intensities (~795 – 820 nm) spectral
4.3 Experimental results

Figure 4.22: Seed gain spectra versus backing pressure. Pump pulse parameters: $\tau_0 = 6.5$ ps, $E_1 = 3.5$ J. Corresponding electron plasma densities are $\sim 2.6 \cdot 10^{18}$ cm$^{-3}$ (4.5 bar), $\sim 6 \cdot 10^{18}$ cm$^{-3}$ (10 bar) and $\sim 1.6 \cdot 10^{19}$ cm$^{-3}$ (28 bar).

Figure 4.23: Seed gain spectra versus pump pulse energy. Pump pulse duration $\tau_0 = 6.5$ ps, electron plasma density $\sim 1.6 \cdot 10^{19}$ cm$^{-3}$.

Figure 4.24: Seed gain spectra versus pump chirp. Pump energy is $\sim 3.5$ J and electron plasma density $\sim 1.2 \cdot 10^{19}$ cm$^{-3}$.
gain curves exhibit stronger oscillations than for the high density plasmas. Deleterious effects on SRBS (EPW wavebreaking, Landau damping and detuning) are reduced at lower plasma densities which may explain the relative rise of what is suggested to be spectral gain from SRBS. It should be noted that indeed gain factors between 820 – 850 nm are substantially increased for tenuous plasmas (not shown here) but overall output energies plummet (as was shown in Fig. 4.8). The energy gain increase for higher pressures and change in the spectral gain curve is here assigned to a transition to sc-SBBS dominated amplification.

The effect of pump energy on the spectral gain is shown in Figure 4.23. Higher energies shift the peak gain to lower frequencies accompanied by gain narrowing. Redshifting in sc-SBBS for increased pump energies is evident since $\text{Re}[\omega_{sc}] \sim (I_0)^{1/2}$ (Eq. 2.13, 2.25). Measurements of the transmitted pump spectra for the high energy gain shots show that the pump signal decreases (Fig. 4.25). Both curves are averaged over 3 shots. The curve for the seeded pump (light green) directly corresponds to the high energy gain shots whose spectral gain curve is shown in Figure 4.23 (red). The unseeded pump transmission spectrum (dark green) is recorded for the same pump energy and plasma density but with a blocked seed pulse. It is unclear if the depletion is caused by stimulated backscattering in the nonlinear regime or other plasma related effects. However, it is found from the relative delay scans that the signal reduction of the transmitted pump is higher for delays where energy transfer is high. This proves that the effect is not simply based on a higher transmission caused by the seed pulse pre-ionizing the target.

The asymmetric behaviour at different chirp signs for the same pulse duration (Fig. 4.24) proofs that amplification is not due to weakly coupled Brillouin or Raman scattering [86]. Spectral gain for the -6.5 ps case (blue) resembles the behaviour for tenuous plasmas where a more pronounced role of SRBS is proposed. Negative pump chirps result in an initial interaction of the seed pulse with spectral components that are more favourable for SRBS which may in return repress the sc-SBBS instability in the later stages. It should be noted that the highest energy transfer efficiency is reached for 6.5 ps, followed by 13 ps and -6.5 ps at last.

![Figure 4.25: Pump output spectra for an unseeded pump pulse traversing the ~1.6 $\cdot$ 10^{19} cm$^{-3}$ target and for the pump interacting with a seed pulse with concurrent high energy gain outputs.](image)
4.3.5 Comparison to 1D PIC simulations

Based on the experimental parameters one-dimensional particle-in-cell (PIC) simulations are performed by colleague Dr. Gregory Vieux (Strathclyde University) using the OSIRIS PIC code. The initial temporal pulse shapes are Gaussian-like profiles (but reach zero power). Spatial profiles of both pulses are Gaussian-like with a flat-top region at the center. The seed pulse propagates from right to left and starts to interact with the leading edge of the pump when entering the plasma. The gas jet target has a 200 µm ramp on both sides and a 2.6 mm density plateau. A resolution of 50 cells per wavelength with 64 electrons per cell is used. In order to investigate the contribution from SRBS to the measured gain spectra the ion mass is set to infinity.

At first, let us take a look at the PIC simulations for seed amplification in a comparably tenuous plasma \( n_e = 5 \cdot 10^{18} \text{ cm}^{-3} \) and at a comparably low pump intensity \( \sim 4 \cdot 10^{15} \text{ Wcm}^{-2} \) (6.5 ps, 1 J). For a positively chirped pump the spectral gain stretches from 790 – 860 nm with a spectral peak intensity at 850 nm (Fig. 4.26). Output pulse duration of the main spike is \( \sim 25 \text{ fs} \). For a negatively chirped pump the peak intensity of the spectral gain is blueshifted (Fig. 4.27) and the output pulse duration is \( \sim 50 \text{ fs} \). Subsequently, at low plasma densities and low pump intensities, SRBS amplification in the initial seed wavelength window is possible while the seed experiences significant pulse compression during the amplification process. Yampolsky et al. showed that the effect of detuning can be reduced by short seed pulse durations, steep seed front slopes and intense pump pulses [24], which gives an explanation why SRBS amplification can be exhibited despite large frequency detuning.

It is found that with increasing plasma density or pump energy amplified spectra shift to lower frequencies. An example is shown in Figure 4.28 where the pump intensity is set to \( \sim 10^{16} \text{ Wcm}^{-2} \) (6.5 ps, \( \sim 3.5 \text{ J} \)) and the plasma density is \( \sim 10^{19} \text{ cm}^{-3} \). The main peak of the amplified spectrum for the positive (negative) pump chirp is located at 870 nm (835 nm). Because of equal pulse and target parameters, aforementioned simulated spectra can be directly compared to the gain spectra measured for \( |\tau_0| = 6.5 \text{ ps} \) in Figure 4.24 (blue and yellow curve). The predominant peak around 815 nm is not remotely reproduced by the simulations and measured pulse durations do not exhibit the compression ratios shown by the simulations (see following chapter). Though it has to be noted that exact pulse duration values are difficult to evaluate from the FROG signals since spontaneously backscattered pump and amplified seed pulse overlap and the seed signal is not always extracted by the pick-up mirror because of spatial shot-to-shot variations. Additionally, simulated output powers are similar for both pump chirps but corresponding experimental data suggests a reduction of the output power by approximately a factor of 5.
Since the simulations for immobile ions (only exhibiting SRBS) are not able to reproduce the measured seed amplification characteristics (especially at high pump intensities and target densities), it is suggested that the main mechanism responsible for the energy transfer from pump to seed is due to the quasi ion mode based sc-SBBS process. A possible explanation for the lack of the strong Raman peaks exhibited in the simulations is the suppression of Raman backscattering when amplification takes place in the sc-SBBS regime [40]. Further simulations for mobile ions are currently in progress.

Figure 4.26: 1D PIC simulation for a positively chirped, medium intensity pump pulse (6.5 ps, ~1 J) interacting with a 150 fs, 15 mJ seed pulse in a 0.005n\textsubscript{c} plasma. The temporal profile of the amplified seed pulse (left) exhibits a pulse duration of ~25 fs in the main spike. The corresponding spectrum (right) shows spectral gain between 790 – 870 nm.

Figure 4.27: 1D PIC simulation for a negatively chirped, medium intensity pump pulse (6.5 ps, ~1 J) interacting with a 150 fs, 15 mJ seed pulse in a 0.005n\textsubscript{c} plasma. The temporal profile of the amplified seed pulse (left) exhibits a pulse duration of ~50 fs in the main spike. The corresponding spectrum (right) shows comparably strong spectral gain around the initial seed wavelength (green curve).
4.3 Experimental results

4.3.6 Characterization of high energy transfer shots

This section presents single shots selected due to their high energy gain at different pump or plasma parameters. The total output spectra (transm. seed and spontaneous backscattering not subtracted), energy gain, seed profiles and finally two corresponding temporal traces are depicted. Tables above the spectra give the pump pulse duration, backing pressure and measured energies for the spontaneous backscattering, transmitted seed, amplified output energy and subsequent energy gain. Single shots with highest energy output are selected, equally only the maximum value from several unseeded pump shots is used for the calculation of the corresponding energy gain. All selected shots are acquired at maximum pump energy (up to 3.5 J) and comparably high plasma densities (~10\(^{19}\) cm\(^{-3}\)). The seed pulse durations is 150 fs and the initial pulse energy is around 15 mJ. Some shots lack a nearfield profile because the diagnostic was not implemented from the beginning of the beamtime. Additionally, it was only possible to surpass the 20 bar backing pressure limit few days into the beamtime which is the reason why no data for the maximum plasma density exists for \(\tau_0 = -6.5\).

Figure 4.30 depicts the two shots with the highest energy gain obtained at an electron plasma density \(n_e \approx 1.2 \cdot 10^{19} \text{ cm}^{-3}\) (0.007\(n_c\)) and pump duration \(|\tau_0| = 6.5\) ps. Highest energy gain for the positive pump chirp is 44 mJ while amplification for the opposite chirp sign yields mere 8 mJ, again showing low efficiency for negative pump chirps.
Energy gain can be further increased when going to higher plasma densities $n_e \approx 1.6 \cdot 10^{19} \text{ cm}^{-3}$ ($0.009 n_c$) as shown in Figure 4.31. Here the highest energy gain is calculated as being 62 mJ which is the best shot of the beamtime. For this particular shot, the pump energy on target is an estimated $\sim 3.2 \text{ J}$ which yields an efficiency around 1.9 %. This is in the realm of other experimental publications on sc-SBBS showing efficiencies between 0.5 – 2.5 % [40, 92, 93]. The current record for the energy transfer ($\sim 45 \text{ mJ}$ [92]) is exceeded by $\sim 35 \%$ and output powers are increased by roughly a factor of 6 due to shorter seed pulse durations.

In addition, Figure 4.31 presents a 48 mJ energy gain shot (different measurement series, similar parameters) since the picture of the nearfield profile is not saturated and it is possible to evaluate the acquired FROG trace.

In general, from the nearfield pictures it is found that the amplified beam is located inside the seed profile but is smaller (due to the smaller pump focus diameter). Spatial shot-to-shot variations can prevent the pick-up of the amplified beam and subsequent FROG measurement. For the 48 mJ shot e.g. it is clearly seen that the pick-up mirror (dark area at the top of the nearfield profile) extracts a part of the amplified signal.

The farfield profiles show some deviations from the low $M^2$ profile for vacuum seed shots. However, the majority of the signal is concentrated in a Gaussian-like focal spot ensuring the applicability of the amplified pulses for further use.

![Figure 4.30: Total output spectrum and farfield profile of a seed pulse interacting with a negatively (left) and positively chirped (right) 6.5 ps, ~3.5 J pump pulse in a $n_e = 0.007n_c$ plasma.](image)

![Figure 4.29: Total output spectrum, farfield and nearfield profile of a seed pulse interacting with a $\tau_0 = 13$ ps, ~3.5 J pump pulse in a $n_e = 0.007n_c$ plasma.](image)
4.3 Experimental results

Figure 4.31: Total output spectrum, farfield and nearfield profile of a seed pulse interacting with positively chirped 6.5 ps, ~3.5 J pump pulses in a $n_e = 0.009n_c$ plasma. Nearfield intensities not to scale since additional ND filters were implemented.

The temporal traces acquired by the FROG diagnostic show a compression of the amplified seed by up to a factor of $\sim$2 when considering the first spike of the output pulse (Fig. 4.32). Seed compression expected from SRBS amplification, as given by the PIC simulations in chapter 4.3.5, is well below 50 fs for comparable parameters. Evaluable FROG traces are rare and show strong shot-to-shot deviations (typical values are between 70 – 200 fs) but the high pulse compression predicted by the nonlinear SRBS regime are not observed experimentally. This should be another indicator that the amplification process at high pump intensities and target densities can not be explained by a model that ignores the plasma ion motion and therefore possible stimulated Brillouin backscattering processes. The temporal structure of the amplified seed pulse is reminiscent of the $\pi$-pulse structures that appear in case of nonlinear stimulated backscattering processes. First proposed and measured for nonlinear SRBS, it was recently shown that the $\pi$-pulse solution also applies for stimulated Brillouin back scattering in the nonlinear regime [107]. However, this is no definite proof that a nonlinear regime is accessed since strong pump chirps can equally lead to temporal seed oscillations in linear regimes [108].

Figure 4.32: Temporal profile of the amplified seed pulse for the $E_{\text{gain}} = 48 \text{ mJ}$ shot in Figure 4.31 (left) and for a shot with comparable gain and equal parameters as shown in Figure 4.29 (right).
4.4 Summary

Record energy transfers from long, high energy pump to short multi-millijoule seed pulses by plasma-based parametric amplification are presented. Energy gain for the best shot is around 62 mJ exceeding the previous record for plasma-based parametric amplification by ~35% [92]. Due to comparably short seed pulse durations and additional pulse compression, output powers are multiples of current record output powers. Beam quality of the output seed pulses is significantly improved compared to aforementioned publication that showed a strongly fragmented beam after amplification. This is likely due to the fact that plasma densities are one order of magnitude lower, equally reducing the growth rate for filamentation and or the short seed pulse durations allowing for the amplification process to take part in the undisturbed front of the pump pulse (as proposed in [92]).

Two indicators of amplification in the nonlinear regime are found, namely a decrease of the pump output signal that is only observed for the high energy gain shots and the temporal structure of the output seed pulses exhibiting (π-pulse like) oscillations.

It is suggested that the main amplification mechanism at high pump intensities (~10^{16} W cm^{-2}) and high plasma densities (~10^{19} cm^{-3}) is the strongly coupled Brillouin backscattering instability. Though no final evidence can be provided, several arguments to justify this assumption are made:
1. Pump and plasma parameters are above the theoretical threshold for the onset of sc-SBBS [37].
2. Experimental results show high energy transfer from pump to seed for stimulated Brillouin backscattering, while stimulated Raman backscattering reaches higher efficiencies and compression ratios [40, 92, 93, 95].
3. Spectral gain curves show highest amplification around the initial central wavelength of the seed pulse which is only a few nanometers shifted from the central wavelength of the pump pulse. SRBS typically requires much larger shifts, especially at the high plasma densities utilized in this experiment. Super broadband amplification by superradiant Raman amplification is excluded since the SRA threshold is not surpassed and it was shown that below threshold SRA is highly unlikely [35].
4. Spectral gain peaks are very robust to changing pressures or relative pulse delays. The three-wave interaction of two pulses with an ion or quasi ion mode is significantly less prone to density variations due to the much lower ion frequencies (as compared to EPWs) in relation to laser frequencies. Further, it will be shown in chapter 5 that experimental results from the SRBS campaign shows delay and pressure dependent shifts over tens of nanometers.
5. PIC simulations with immobile ions are not able to reproduce the gain spectra exhibited for the high energy gain shots. Output pulse durations in the simulations are consistently below 50 fs (which would be reached by nonlinear SRBS) while FROG measurements fluctuate between 70 – 200 fs.
4 Parametric backscattering of joule-level pump pulses in sub-quarter-critical plasmas
This chapter presents the results of the stimulated Raman backscattering campaign performed at the Jeti40 laser.

Red shaded area in Figure 5.1 shows where the experiment is located as determined by pulse and plasma parameters. For the most part the wavebreaking threshold for the electron plasma wave is surpassed. Though pulse durations of the SPM shifted seed are below the plasma period, seed intensities are not sufficient to enter the coherent wavebreaking regime [55]. Therefore, wavebreaking is expected to reduce energy transfer from pump to seed [32].

The threshold for strongly coupled Brillouin backscattering is also surpassed. However, experimental results do not indicate participation of Brillouin instabilities to the amplification process. As opposed to chapter 4 where sc-SBBS is proposed as the main amplification mechanism, here pump intensities are about one order of magnitude lower. Subsequently, the experimental parameter range shifts much closer to the sc-SBBS threshold (as given by Eq. 2.23) which is not a hard threshold but defines a soft transition from weak to strong coupling. Another major difference to the previously shown experiment is the presence of seed frequencies satisfying the SRBS frequency matching condition, lower seed intensities and an even shorter pump pulse duration. Focus of the beamtime is laid on the SRBS instability in a well-defined density gradient created by a trapezoid orifice of the gas nozzle. As discussed in previous chapters, chirping the
pump pulse helps to reduce unwanted instabilities like spontaneous Raman forward- and backscattering. At the same time pump chirps lead to increased detuning which can be described by the detuning parameter \( q \) (Eq. 2.17). Since \( q \sim \frac{\partial \omega_{pe}}{\partial x} - 2 \frac{\partial \omega_0}{\partial x} \), the second term describing the chirp of the pump pulse can be compensated by the introduction of a plasma density gradient. Same sign gradients reduce and opposite signs increase the detuning growth rate. The amplification process should therefore be improved e.g. in case of a negative density gradient (as seen by the pump) combined with a positive pump chirp. Both cases \((\pm \frac{\partial \omega_{pe}}{\partial x})\) are investigated at otherwise equal parameters by a simple 180° rotation of the trapezoid nozzle. The results can be directly compared to a common flat-top target that was also implemented during the beamtime. Both gas nozzles are characterized via tomographic interferometry which gives spatially resolved results for the target densities at various backing pressures. This allows for a precise definition of the density gradient as well as peak densities exhibited by the nozzles. In other experimental publications on SRBS in density gradients, ionizations steps or oblique irradiation in pre-ionized plasma channels are used [95, 99]. Those methods complicate the characterization of the plasma gradient and introduce uncertainties concerning the (pulse delay dependent) actual gradient profile in the overlap region.

Figure 5.1: Threshold conditions for different regimes. Red area represents pump pulse and target density parameters utilized during the beamtime. For the threshold calculations the laser wavelength \( \lambda_0 \approx 800 \text{ nm} \), seed intensity \( 1 \cdot 10^{14} \text{ Wcm}^{-2} (a_1 \approx 0.0048) \) and quasi-neutral \( \text{H}_2^{2+} \) plasma are used.
5.1 Jeti40 laser system

Jeti40 is a semi-commercial, high intensity, CPA laser system with a peak power of 40 TW. The fs-oscillator generates a pulse train of 12 fs pulses at an average power of 300 – 400 mW. In a first step, the pulses coming from the oscillator are amplified to 5 mJ and the repetition rate is reduced to 10 Hz. Pulse duration is increased to ~800 ps by the stretcher to stay below the damage thresholds of the Ti:sa amplifier crystals. After passing through a chain of multipass amplifiers pumped by frequency doubled ND:YAG lasers, energy per pulse is ~1.3 J. The beam diameter is widened to d = 60 mm in a 1:5-telescope before being sent to the grating compressor. Here the \( \lambda_c = 800 \text{ nm}, \Delta \lambda \approx 60 \text{ nm} \) pulses are compressed down to \( \tau = 25 - 30 \text{ fs} \). Compressor efficiency is ~60% resulting in a final pulse energy of \( E \approx 0.8 \text{ J} \).

5.2 Setup and diagnostics

A schematic of the setup is found in Figure 5.3. To obtain two different pulses that will be utilized as pump and seed a 85:15-beamsplitter is placed shortly behind the compressor inside the vacuum beamline. The reflected (pump) pulse follows the ordinary beamline to the \( d = 85 \text{ cm} \) target chamber. A dielectric mirror inside the chamber sends the 60 mm beam to the ~2.5 m long target chamber extension. Here a f = 3000 mm dielectric spherical mirror focuses the beam onto the gas jet target. Both mirrors are motorized appropriately to optimize the focus profile and position. A slightly tilted vacuum compressor grating (which is noticed only after the beamtime) and the non-zero degree angle on the spherical mirror yield a focal spot that requires significant corrections.

![Figure 5.2: Schematic of the final seed pulse extraction setup.](image)
by the adaptive mirror which is implemented in the laser beamline. The pulse that is transmitted by
the 85:15-beamsplitter will finally serve as the seed and probe pulse in the experiment. The
transmitted pulse exits the vacuum beamline, is apertured down to ~20 mm and subsequently
compressed by an additional double-pass grating compressor. The pulse is then focused onto the
entrance of the hollow-core fiber by an f = 1000 mm lens. The HCF is fixed inside the gas cell by a
v-shaped metal frame. The 2 m gas cell has thin (~1 mm) entrance and exit windows that are tilted
at Brewster's angle. Behind the gas cell the SPM broadened pulse is collimated and compressed by
a two pairs of chirped mirrors. Since the chirped mirror setup has a fixed group velocity dispersion
(GVD), a pair of motorized wedges introduces a variable dispersion length to ensure the shortest
possible pulse duration.

Before entering the target chamber a part of the seed pulse is separated by a pellicle beamsplitter to
generate a probe pulse that is used for the shadowgraphy diagnostic. Two delay stages are
implemented, one in the seed arm to adjust the timing relative to the arrival of the pump pulse at
the target position and one in the probe arm to adjust the timing relative to seed and pump. The
probe pulse passes several mirrors and traverses the target perpendicular to the other two pulses.
The seed is focused by an f = 1000 mm dielectric spherical mirror onto the gas jet target.

The amplified seed (respectively SRBS signal) is at first collected 3 m behind target by a 4“ gold-
coated mirror (45° angle of incidence) that is placed in the extension box of the pump focusing
optic. The pulse exits the vacuum and is collimated shortly before entering the diagnostics table.
The output pulse extraction setup is later modified, by setting up two 2“ squared mirrors 0.5 m
behind the target and early collimation inside vacuum (Fig. 5.2). This modification was necessary
due to uncharacteristic wide backscattering angles.

The diagnostics for the SRBS signal are set up on multiple breadboards that are served by a
successive line of 2“ beamsplitters. The diagnostics include two silica CCD-cameras as near- and
far-field diagnostic, a Coherent EnergyMax pyroelectric energy sensor and an Andor
Shamrock193i spectrograph with an Andor Newton 920 CCD camera attached. The spectrograph is
calibrated by a krypton lamp shortly before the start of the beamtime.
5.2 Setup and diagnostics

Figure 5.3: Schematic of the experimental setup.
5.2.1 Ultrashort seed pulse generation by self-phase modulation in a gas-filled hollow-core fiber

The seed pulse in this experiment is generated by self-phase modulation (SPM) in a gas-filled hollow-core fiber (HCF) with subsequent compression. The HCF serves as the waveguide for the pulse ensuring long interaction lengths with the working gas at a high intensity (similar to Bessel beam focusing by axicon). Compared to solid-core fibers the hollow-core structure allows for much higher damage thresholds. The guiding mechanism is based on a photonic bandgap since guiding by refractive index is only possible for propagation in a medium with a higher refractive index than the cladding.

The laser pulse is coupled into the fiber by focusing it onto the entrance of the $d = 200 \mu m$ hollow-core. While traversing the fiber core, the intensity induces a nonlinearity of the refractive index of the respective working gas (Eq. 3.8). The time-dependent phase shift $\Phi_{NL}$ (Eq. 3.9) changes the instantaneous frequency $\omega(t) = \omega_0 + \frac{\partial \phi_{NL}(t)}{\partial t}$. The pulse spectrum is modified whereas the rising (declining) slope of the pulse experiences a self-induced downshift (upshift) in frequency. Assuming a positive or non-existent pulse chirp and a positive nonlinear refractive index $n_2$, SPM leads to an increase of the spectral width. Whereas pure SPM (not taking into account dispersion of the medium) has no effect on the temporal profile of the pulse. Related to this experiment, spectral modulation is supposed to create new low frequency components that satisfy the frequency condition for SRBS. At the same time the broadened spectra support ultrashort pulse durations which increase the pulse power to feasible values for SRBS seeding. Neon and argon are tested as working gases at different pressures and input energies. Gas cell pressures reach up to 700 mbar and input pulse energies are typically ~1 mJ. Higher pulse energies lead to unwanted ionization and damage the optical fiber, hence degrading energy transmission. In this experiment energy transmission is around 50% for optimized coupling. Optimization is done by finding the correct longitudinal focal position (by linear stage of the lens) and fiber orientation (by two linear xy-stages at the two ends of the gas cell). Ideally, a close to perfect Gaussian mode is exhibited after the fiber exit. It should be noted that energy transmission can be up to 80% for a better spotsize-to-core area. However, geometrical restrictions did not allow an improvement for this setup. Input pulse durations are adjusted accordingly to maximize spectral width. This is done on-shot by recording the output spectra with an OceanOptics4000 fiber spectrometer while varying the compressor grating distance. Best results are obtained for short pulses (few tens of femtoseconds) with a small chirp compensating for group velocity dispersion inside the gas-filled HCF.
5.2 Setup and diagnostics

Figure 5.6 shows the spectra of the SPM broadened pulses in argon and neon. The input pulse spectrum can be found in Figure 5.7. All spectra show strong oscillations of the spectral intensity. Given a normal Gaussian or sech\(^2\) pulse shape, the shift of the instantaneous frequency inherits the same value at two distinct times which will lead to the generation of two similar frequency components that can interfere. The interference of those frequency components leads to the oscillating spectral intensity exhibited here. Best results in terms of broadening and generating comparably intense, low frequency components are achieved in argon. Total output energy measured at the fiber exit is 500 µJ. Around 25% of the energy is in the wavelength region above 825 nm which is ~130 µJ. Due to losses from the numerous optical elements before hitting the target this value is then reduced to ~45 µJ.

In the next step the broadband pulses are compressed to the bandwidth limit. A total of 16 bounces over 2 pairs of chirped mirrors introduce a GVD that overcompensates the chirp acquired in the hollow-core fiber. The counteracting normal dispersion from the different transmission optics is not sufficient to remove this chirp. Additionally, a set of motorized fused silica wedges introduce a variable group delay dispersion that allows for shortest pulse durations on target. Measurements of \(\tau_1\) are performed by a FastLight Whizzler. The results for an optimized propagation length in the wedges are shown in Figure 5.4. It should be noted that the Whizzler filters out wavelengths that are outside the acceptance range of 700 – 900 nm. Additionally, a systematic error leads to an 10 – 100 fold increase in the recorded shot numbers above 30 fs (red shaded bars). So the shortest measured pulse duration for the broadband seed pulses generated in argon is therefore estimated to be ~18 fs but might be a few femtoseconds shorter because of the cut-off at 700 nm. A picture of a corresponding typical nearfield profile is shown in Figure 5.5. Unfortunately it was not possible to extract a Gaussian mode (LP01) but only signals with a higher mode index. Once adjusted, spectral and mode stability are appropriate for a few hours (~4 h) of SRBS data acquisition until the spatial drift from the laser requires readjustment.

Figure 5.4: Pulse duration of the SPM broadened seed measured by a FastLight Whizzler behind the chirped mirror setup.

Figure 5.5: Nearfield seed profile after the HCF.
5.2.2 Seed and pump pulse parameters

Pump pulse durations, measured by an Amplitude Sequoia 800, are kept at a constant value of \( \tau_0 = 3.6 \text{ ps} \) (Fig. 5.9). The third-order autocorrelation measurement shows a significant prepulse with a peak intensity \( I_{pp} = \frac{1}{4} \cdot I_0 \) arriving \( \sim 7.5 \text{ ps} \) before the maximum of the main pulse. Hence, pre-heating and pre-ionization of the target have to be taken into account e.g. in order to match experimental results with simulations. The laser spectrum is shown in Figure 5.7. Central wavelength is around 800 nm with a FWHM bandwidth of \( \Delta \lambda \approx 60 \text{ nm} \), spectral peak intensity of the asymmetric spectrum is around 820 nm.

The seed pulse duration is at an estimated \( \tau_1 = 18 \text{ fs} \). The typical seed spectrum at target is shown in Figure 5.8 (averaged over 3 shots). The central wavelength is at \( \lambda_c = 806 \text{ nm} \) while the important redshifted wavelength components stretch to \( \sim 905 \text{ nm} \) where the noise level of the spectrograph is reached.

The f-numbers are f/50 (pump) and f/85 (seed). Aberrations (from the tilted compressor grating) result in an energy of only \( \sim 175 \text{ mJ} \) (about 30\% of the \( E_{\text{total}} \)) contained in the pump focus. The seed contains an energy of up to 170 \( \mu \text{J} \) but is spread out over several focus diameters compared to the pump. Estimated peak intensities are \( I_0 = 1 \cdot 10^{15} \) and \( I_1 = 1 \cdot 10^{14} \text{ Wcm}^{-2} \) for the pump, respectively seed pulse.
5.2 Setup and diagnostics

5.2.3 Spatial and temporal overlap

During this beamtime it was not possible to find the temporal overlap of the seed and pump pulse by shadowgraphy (as described in chapt. 4.2.2) due to insufficient seed intensities. Instead a temporal overlap diagnostic is implemented that utilizes intensity cross-correlation in a nonlinear crystal (Fig. 5.10). Spatial overlap is ensured by the focus diagnostics imaging a needle tip where pulse overlap is established by the motorized mirrors and focusing optics.
5.2.4 Gas jet targets

The target is a gas jet generated by either a conical 1.5 mm or an isosceles trapezoid orifice (dimensions: a = 1 mm, b = 3 mm, h = 3 mm). The valve is triggered 2.5 ms before arrival of the pulses and has an opening time of 3 ms.

The working gas is molecular hydrogen with typical applied backing pressures p = 6 – 13 bar, resulting in electron densities of $2 \times 4 \times 10^{19}$ cm$^{-3}$ for the conical nozzle and an average density of $9 \times 10^{18} – 2 \times 10^{19}$ cm$^{-3}$ for the trapezoid nozzle (fully ionized H$_2$, 0.5 mm beam height above nozzle). In the commonly used description of the target density in terms of critical density the range $0.005n_c < n_e < 0.021n_c$ is covered. The exact height above nozzle can be determined by the calibrated shadowgraphy camera recording the density disturbances caused by the pump pulse. The characterization of the two gas nozzle shapes (conical, trapezoid) is done by tomographic interferometric measurements of the neutral gas density. The results of those measurements, converted to electron densities in fully ionized H$_2$ gas, are shown in Figure 5.11 and 5.12. The labels normal and inverse in Figure 5.12 denote the direction of measurement, sum is the average of those two curves. Densities generated by the trapezoid nozzle are lower for similar backing pressures mainly due to the increased orifice surface compared to the conical nozzle. The density gradient near the center of the trapezoid nozzle is an estimated $-3 \times 10^{18}$ cm$^{-3}$mm$^{-1}$ which here corresponds to $\left| \frac{\partial \omega_{pe}}{\partial z} \right| = \left| \frac{2.03 \times 10^{14}s^{-1} - 1.78 \times 10^{14}s^{-1}}{1 \text{ mm}} \right| \approx 2.5 \times 10^{13} \frac{1}{\text{s mm}}$. 

![Schematic of the temporal overlap diagnostic based on intensity cross-correlation.](image)
5.2 Setup and diagnostics

Figure 5.11: Electron density profile of the 1.5 mm conical nozzle at backing pressures of 6, 8 and 12 bar.

Figure 5.12: Electron density profile generated by the trapezoid nozzle at 8 bar backing pressure. Added picture of the gas jet nozzle with respective beam directions for the negative ramp profile as seen by the pump beam.
5.3 Experimental results

In the following the acquired data of the Jeti40 SRBS campaign is presented. Central aspect is the comparison of the two target nozzles including opposite density gradients obtained by a rotation of the trapezoid nozzle.

The first section shows the effect of the relative pulse delay on the output spectra and efficiency of the SRBS instability. Delay dependent wavelength shifts of the Raman peaks are observed that can be explained by the pump chirp and overlap position inside the plasma. The next section compares detuning growth rates of the negative and positive density gradient. The large pump chirp rate is not remotely compensated by the introduced density gradient. However, a significant reduction of seed output energies is obtained when using the gradient which increases the detuning growth rate. Furthermore, results for the conical and trapezoid nozzle at different electron plasma densities show that SRBS efficiency is improved (in absence of filamentation) for pump to plasma frequency ratios lower than previously suggested by PIC simulations [41]. Chapter 5.3.3 presents 1D PIC simulations (courtesy of G. Lehmann, Düsseldorf university) for the negative density gradient that are in good agreement with experimental data. Therefore, conclusions about the pulse duration and overall temporal structure of the amplified pulse can be drawn. The last two sections treat the amplification of an unshifted seed pulse and show a collection of the best shots in terms of energy output including a discussion of the energy gain and efficiency.

Seed energy gain $E_{\text{gain}}$ is defined as described in chapter 4.3. The total measured energy is subtracted by the backscattered energy of the pump for blocked seed pulses and the transmitted seed energy. Here the transmitted energy with a blocked pump pulse is equal to the initial seed energy since intensities are too low to ionize the target, no ionization-, plasma heating- or scattering losses occur. For the efficiency calculation (chapt. 5.3.5) the transmitted energy through the ionized target is considered.

Vibrations of the building lead to strong spatial variations that are especially significant in the pump arm since the HCF in the seed arm partly compensates for the spatial jitter. The deviation of the pump focus amounted in the worst case to more than a pump focus diameter. The strong spatial deviations in combination with small shot-to-shot seed fluctuations from the broadening process led to strong fluctuations of the acquired data which was tried to compensate for by performing at least 20 shots per parameter setting.

All plotted spectra (acquired by the Andor spectrograph) are corrected in terms of background, CCD quantum efficiency, neutral density filters and optical components inside the spectrograph including a gold grating and several aluminum mirrors (Fig. 5.13).
5.3 Experimental results

5.3.1 Frequency shift and output energy dependence on relative pulse delays

Since the pump pulse is stretched to several picoseconds and exhibits a linear chirp, the overlap position not only determines the interaction length but should also affect frequency resonance. A schematic is shown in Figure 5.14. Gray represents the target area defined by the gas jet diameter and red indicates the interaction length of the two pulses where the plasma wave is driven by the beat. Here the displayed pump pulse is positively chirped, hence an early seed pulse only interacts with low pump frequencies and a late the seed overlaps with the high frequency components. For all 3 delay scans presented in the following (conical, trapezoid, reversed trapezoid) the three shots with the highest energy output for every delay value are selected and averaged for the spectral and energy gain plots. The delay scan for the 1.5 mm conical nozzle is shown in Figure 5.15. The output seed spectrum is attenuated due to the
interaction with the target plasma. Even at the zero relative delay (\( \approx \) shortest seed delay of the measurement series) where no amplification is observed either the prepulse of the pump arriving \(-4 \) ps before the main pulse and or the very leading edge of the main pulse already generate a plasma mitigating seed transmission. The energy loss in the \( n_e = 2.2 \cdot 10^{19} \) cm\(^{-3} \) plasma is around 10 – 20\% as determined by pyroelectric energy sensor measurements. The reduction may be overrated in the spectral curves since plasma effects lead to a spread and or directional change of the seed beam which then might be not fully transmitted by the entrance slit of the spectrograph.

The SRBS spectra are located between 875 – 935 nm. A delay dependent shift of the spectral peak intensity is found. For small relative delays, the seed pulse traverses the target and only interacts with the rising slope and subsequently the low frequency components of the pump. Output spectra are therefore equally shifted to longer wavelengths as determined by SRBS frequency matching. Increasing relative delays lead to a complete seed pump overlap in the plasma (proper overlap in Fig. 5.14) which maximizes energy gain as it is observed here for relative delays around \(-3 \) ps. The 3 ps delay most likely corresponds to the case where the seed pulse meets the front of the pump pulse near the seed entrance to the target. An increase to \( t_d = 4.2 \) ps further shifts the Raman peak to shorter wavelengths, corresponding to the case of a late seed in the previously shown schematic where the seed only interacts with the high frequencies of the pump. The overall frequency range covered by various delays is comparable to the FWHM bandwidth of the pump pulse. Double peak structures of the Raman backscattering spectra do not only originate from the averaging process.

![Figure 5.15: Delay dependent output spectra and energy gain for the 1.5 mm conical nozzle, electron plasma density \( n_e = 2.2 \cdot 10^{19} \) cm\(^{-3} \).](image-url)
5.3 Experimental results

since different shots show either a solitary peak at a distinct wavelength (e.g. 910 and 925 nm, yellow curve) or the double peak structure simultaneously at both wavelengths. Results for the delay scan with the isosceles trapezoid nozzle oriented with the short base to the pump beam (→ negative density gradient as seen by the pump) are shown in Figure 5.16. The average electron density is \( n_e = 1.2 \cdot 10^{19} \text{ cm}^{-3} \) and therefore lower than in the previously shown data from the conical nozzle. This explains shorter central wavelengths of the Raman peaks and the higher transmission of the initial seed spectrum. The zero delay is again arbitrary set to the earliest seed pulse arrival, precise comparison to Figure 5.15 is not possible due to the positional uncertainties introduced by the nozzle exchange procedure though it was tried to match the nozzle positions with the help of the sideview diagnostic. The delay range for high energy gain is increased due to the doubling of the target length to 3 mm. Again, a delay dependent spectral shift of the Raman peak is observed. The broad (double peak) spectra for the late seed pulses (\( t_d = 4.2 \) and 5 ps) are exhibited consistently also at single shot spectra. Compared to the conical nozzle, the negative density gradient gives broader output spectra at delays that ensure optimized pulse overlap. In terms of seed energy gain, the conical nozzle seems to be superior to the ramp target. However, a direct comparison of both targets (negative gradient vs. flat-top) is not unambiguous due to different plasma densities, target lengths and backscattering cone angles (see chapt. 5.3.5).

![Figure 5.16: Delay dependent output spectra and energy gain for the 3 mm trapezoid nozzle in the negative gradient orientation (narrow side to pump), average electron plasma density \( n_e = 1.2 \cdot 10^{19} \text{ cm}^{-3} \) (0.007\( n_c \)).](image-url)
The results for the delay scan with the trapezoid nozzle rotated by 180° (→ positive density gradient as seen by the pump) show strongly reduced seed amplification (Fig. 5.17). Relative delay values are consistent to Figure 5.16.

A more thorough discussion of the characteristics exhibited by the opposite sign density gradients and a comparison to the flat-top nozzle is found in the following chapter.

Figure 5.17: Delay dependent output spectra and energy gain for the 3 mm trapezoid nozzle in the positive gradient orientation (broad side to pump), average electron plasma density \( n_e = 1.2 \times 10^{19} \text{ cm}^{-3} \) (0.007\( n_c \)).

5.3.2 Detuning growth rates in opposite sign density gradients and comparison to a flat-top target

The density gradient near the center of the trapezoid nozzle is \( \gamma \approx 2.5 \cdot 10^6 \). The pump chirp can be approximated as linear and from the FWHM bandwidth of ~60 nm (\( \lambda_c \approx 800 \text{ nm} \)) and the pulse duration of 3.6 ps one can estimate the frequency gradient of the pump as

\[
\frac{\partial \omega_0}{\partial z} = \frac{2.44 \cdot 10^{15} \text{s}^{-1} - 2.27 \cdot 10^{15} \text{s}^{-1}}{3.6 \cdot 10^{12} \text{s} \cdot \text{c}} \approx 1.57 \cdot 10^{14} \frac{1}{\text{s} \cdot \text{mm}}.
\]

The large pump chirp rate is expected to
significantly reduce spontaneous backscattering from the pump but leads to frequency detuning that can reduce SRBS efficiency at the same time. It is proposed to introduce plasma gradients compensating for pump chirps so that resonance remains constant [24]. Equation 2.17 shows that equal signs for the gradients can compensate each other leading to a reduced or zero detuning growth rate in the backscatter direction. Since the pump has a fixed positive chirp, compensation by a plasma gradient is ensured in case of a negative density gradient (as seen by the pump pulse). The density gradient is not sufficient to fully compensate for the pump chirp, one calculates the dimensionless detuning growth rate introduced by the pump chirp as

$$q_0 = \frac{4c(\partial \omega_0)}{a_0^2 \omega_0 \omega_{pe}} \approx 1.61$$

which can be reduced (negative gradient) or increased (positive gradient) by

$$q_{pe} = \frac{2c(\partial \omega_{pe})}{a_0^2 \omega_0 \omega_{pe}} \approx 0.13.$$ 

All calculated values are for $n_e \approx 1.2 \cdot 10^{19}$ cm$^{-3}$, $I_0 = 1 \cdot 10^{15}$ W cm$^{-2}$ and $\lambda_c = 800$ nm. The reduction or increase of the detuning growth rate is less than 10%. However, the difference in terms of seed amplification is significant. In a continuous measurement series of 260 shots with the negative gradient 5 shots exceeded an total energy output of 650 µJ with a maximum energy of 850 µJ. In the subsequent measurement with the positive gradient the same amount of shots shows only 4 shots exceeding 245 µJ with a maximum total output energy of mere 300 µJ. The vast majority of shots at the positive gradient do not show any sign of amplification. It should be noted that the poor result for $\alpha > 0$ are not due to a lack of pulse overlap since the proven methods for establishing temporal and spatial overlap were implemented after nozzle rotation, followed by additional scans of the pulse delay and seed focus position.

Output spectra for the two gradients are shown in Figure 5.18, for every curve three shots with maximum energy output are selected, averaged and normalized. Average total output energy is 730 µJ for the negative gradient (blue) and 295 µJ for the positive gradient (yellow). It can be argued that the considerable change in seed amplification is caused by the difference in the detuning growth rate since the seed spectrum is sufficiently broad to provide frequency matched components for various pump frequencies.

Spontaneous Raman backscattering (RBS) of the pump is already strongly mitigated by the pump chirp. The chirp rate can be defined as $\alpha = \pm \frac{(\Delta \omega)^2 \delta^2 - 64 \log^2(2)}{2 \omega_0^2 \delta^2}$ which yields $-4.4 \cdot 10^{-6}$ for aforementioned pump parameters [86]. 2D PIC simulations show that spontaneous backscattering is efficiently mitigated for $2\alpha \geq \frac{\gamma_{SRBS}}{\omega_0^2 \delta^2}$ with $\gamma_{SRBS}$ the growth rate of the Raman instability (Eq. 2.12) [106]. The condition is surpassed by more than one order of magnitude which explains comparably weak RBS signals. However, it should be noted that the most intense part of
the spontaneous pump backscattering is not reflected by the squared mirror setup and therefore not acquired by the diagnostics.

Experimental data shows that the positive gradient is, besides decreasing seed amplification, further lowering RBS. 40 consecutive shots (with blocked seed pulse) did not trigger the pyroelectric energy sensor since the backscattering from the plasma noise was below threshold which can be converted to energies invariably below 20 µJ. For the negative gradient the corresponding measurement showed only 3 shots below threshold, an average backscattering energy of 35 µJ and a maximum of 150 µJ. Not the full energy difference is accounted for by the spontaneous Raman signal since a decrease of backscattered fundamental pump spectrum is observed at the same time. However, it is possible to show that RBS experiences a distinct reduction if the pump traverses the positive density gradient (Fig. 5.19). Every curve is averaged over 3 unseeded pump shots with highest signal count at the Raman wavelengths. To scale spectrograph images of the corresponding shots are shown for comparison. It is found that the negative density gradient is superior in terms of output energy and bandwidth for SRBS and RBS alike.

Figure 5.18:
Normalized output spectra for the negative (blue) and positive (yellow) plasma gradient target. Average plasma density is \( n_e = 1.2 \cdot 10^{19} \text{ cm}^{-3} \) (0.007\( n_c \)).

Figure 5.19:
Corresponding RBS spectra for both gradients with the to scale spectrograph images of the shots that are used for averaging.
5.3 Experimental results

Let us now compare the flat-top and negative gradient target in terms of amplification characteristics. Figure 5.20 shows the SRBS spectra for the conical nozzle at different plasma densities. Every curve is averaged over 40 shots, dashed vertical lines indicate where the peak of the amplified signal is expected. Electron densities at different pressures follow from the tomographic interferometry measurements (see chapt. 5.2.4). Good agreement is found when calculating with the central frequency of 800 nm for the pump pulse neglecting electron plasma temperature. Corresponding pump to plasma frequency ratios are labelled at the top. The curves are acquired for similar relative delays (coinciding with highest averaged energy output), the 13 bar measurement is for a slightly increased delay (+0.8 ps) which can easily account for a 10 nm blueshift of the SRBS peak as it was shown in chapter 5.3.1. Increasing electron densities decrease the average total output energies from 600 µJ (0.013n_e) to 490 µJ (0.019n_e) and finally 400 µJ. Energy differences are slightly smaller when taking into account lower transmission ratios at higher pressures but the effect is still significant as it is also shown by the integrated spectrograph signal.

SRBS spectra acquired with the negative density gradient are shown in Figure 5.21. Every curve is averaged over 20 shots. Again, for the high backing pressure (13 bar) the delay is ~0.8 ps longer compared to the remaining curves. Dashed vertical lines represent the theoretical Raman peaks as follows from the electron plasma densities at different backing pressures. The given electron density is the averaged value over the whole ramp profile. A better fit to the experimental data is achieved when calculating with the spectral peak intensity of the pump pulse (818 nm) instead of the central frequency. This may suggest that the pump components in the leading edge strongly determine the amplification process and output wavelength before deleterious instabilities (as wavebreaking) start to emerge. Another possible explanation is a significantly higher electron temperature compared to the flat-top profile that would increase the wavelength shift according to Eq. 2.4.

The maximum average output energy is found for 10 bar (0.009n_e) and the minimum for 13 bar (0.012n_e) though statistical errors are large and energy differences too small to give a reliable ranking for all backing pressures. However, the data suggests that (independent from the particular target) best results are obtained for output wavelengths around 900 nm. It is therefore suggested that seed pulse amplification is preferable for $\frac{\omega_p}{\omega_0}$ – ratios that are significantly lower than proposed in [41]. The drop in efficiency when further increasing the electron plasma density may be accounted for by the declining spectral intensity of the seed pulse above 900 nm (see e.g. red dashed curve in Fig. 5.20). In chapter 5.3.4 it will be shown with a proper filter selection in front of the spectrograph that the seed spectrum stretches further than 905 nm and spectral seed components well below the µJ-level can be amplified at ultra-high gain factors.
5.3.3 1D PIC simulation for the negative density gradient profile

One-dimensional PIC simulations for the negative density gradient are performed by G. Lehmann (Düsseldorf University) using the EPOCH PIC code. The initial temporal profile of the pump is modeled by a sequence of Gaussian profiles in order to fit to the profile that was measured by third-order autocorrelation (Fig. 5.9). At first a 1.4 ps prepulse with an intensity of $4 \cdot 10^{14} \text{ W cm}^{-2}$ arrives 7.5 ps before the peak of the main pulse. The 3.6 ps main pulse has a peak intensity of $\sim 1 \cdot 10^{15} \text{ W cm}^{-2}$ and is trailed by a 2.1 ps post-pulse ($3 \cdot 10^{14} \text{ W cm}^{-2}$) arriving 3 ps after the main
5.3 Experimental results

Figure 5.22: Temporal intensity profile of the amplified seed after pulse interaction in the negative density gradient target as given by EPOCH PIC code simulations.

The pump pulse is linearly chirped (according to the experimental parameters) with a central wavelength of 800 nm. Central wavelength of the 18 fs (temporal Gaussian) seed pulse is set to 830 nm. Four linear density ramps are used to model the gradient profile shown in Figure 5.12. The peak electron plasma density is 0.008$n_c$. It should be noted that the target is already fully ionized so that ionization processes affecting the leading edge of the pump pulse are not covered by the simulations. Seed pump interaction starts at the beginning of the density ramp as seen by the seed pulse (position $r = 1$ mm in Fig. 5.12). A resolution of 32 cells per wavelength is chosen, resulting in 300 particles per cell in the 4 mm simulation box.

The temporal intensity profile of the output pulse train in the backscatter direction is shown in Figure 5.22. The leading pulse has a duration of $\sim 75$ fs and contains almost 90% of the total energy. In the first stage of the amplification process the seed pulse duration increases to several hundreds of femtoseconds due to staggered pump backscattering at the EPW. Shortly thereafter exponential growth of the leading pulse front starts and temporal compression down to $\sim 75$ fs is reached within a $\sim 1$ ps time window. Backscattering at the EPW continues after the leading pulse and lasts longer than the initially stretched seed duration resulting in a pulse train $\geq 1$ ps.

The pulse spectrum is computed by a fast Fourier transform (FFT) that can be applied to the simulated electric field of the output pulse. It is found that the simulation is in good agreement with the experimental data acquired at equal parameters. The normalized FFT of the simulated electric field (blue) and the normalized spectrum for the best shot of the measurement series (red, dashed) are shown in Figure 5.23. Additionally, six shots with the highest output energy at corresponding parameters are selected for the averaged spectral curve (yellow, dashed). Biggest deviations between simulation and experimental data are found in the high frequency range ($\lambda = 845 - 860$ nm) where the spectral peak observed in the measurements is much broader and blueshifted compared to the PIC data. Separated FFTs, one for the leading pulse and one for the trailing pulse train, provide an approximation of the backscattered spectral components at different
times. It should be noted that this method is not perfectly accurate since interference effects are not included. It is found that the post-pulses mainly account for the spectral peak at short wavelengths (Fig. 5.24). The blueshift of the secondary peak compared to the central wavelength of the main pulse spectrum can be explained by particle trapping reducing the EPW frequency. Simulations show that the output spectrum between 860 – 905 nm is robust to small changes in plasma density or temperature. The secondary peak at shorter wavelengths strongly depends on those parameters which additionally points to the emergence at later interaction times when plasma conditions are altered by the traversing pump and seed. It is therefore very sensitive to shot-to-shot fluctuations and could also be an indicator for the energy ratio of leading to trailing pulses.

Figure 5.23: Fast Fourier transform of the simulated E-field. Dashed lines show experimental data for the best shot (red) and the average of 6 shots (yellow) with the highest energy gain.

Figure 5.24: Separate FFTs of the simulated E-field for the leading pulse (blue) and the trailing pulses (red).
5.3 Experimental results

5.3.4 High gain factor amplification of unshifted seed pulses

A separate measurement is performed where the hollow-core fiber is evacuated by a scroll pump resulting in a cell pressure $< 10^{-2}$ mbar. No shifting or broadening by the working gas is expected in this case so that the amplification process for a similar pump and seed spectrum can be evaluated. The lack of spectral broadening additionally leads to longer seed pulse durations which are expected to be around 40 fs. The transmitted seed energy is (similar to the case of the gas-filled HCF) $\sim 160 \pm 15 \, \mu J$, spontaneous backscattering energy is $55 \pm 45 \, \mu J$ with the maximum value 210 $\mu J$. Therefore, in the worst case up to 385 $\mu J$ could be measured for the pulse overlap shots without the participation of any amplifying process. The best shot of the series showed a total energy of 650 $\mu J$ which is a clear sign for the amplification of the unshifted seed pulse. Spectra of selected high energy output shots for pulse overlap (blue curves), compared to unseeded pump backscattering (green curves), show significant spectral gain at the Raman wavelengths (Fig. 5.25). After replacing the set of neutral density filters in front of the spectrograph with dielectric longpass filters (cutting below 850 nm) an unpumped seed spectrum is acquired (Fig. 5.26). It is found that the spectrum stretches far to the low frequency side which may be due to weak self-phase modulation in the HCF cladding or lenses and windows traversed by the seed pulse. However, the signal in that frequency range is more than $\sim 10$ orders of magnitude lower compared to the properly shifted seed in a gas-filled HCF. Therefore, amplification with huge spectral gain factors but lower efficiency is achieved. For the same target parameters the seed energy gain is roughly tripled when utilizing the shifted seed pulse. Furthermore, it is concluded that the reduced energy gain cannot be explained by the overall lower seed intensity (due to the longer pulse duration). In a separate measurement series (not shown here), the pulse duration of the shifted seed is consistently increased by introducing a group velocity dispersion (via the motorized fused silica wedges). The subsequent delay in the seed arm is compensated by the motorized delay stage. Pulse durations reach from shortest seed duration ($\sim 18$ fs) up to $\sim 100$ fs when introducing a total of $\sim 1$ cm glass. No significant reduction of the energy gain that can explain the 3 fold decrease exhibited for the unshifted seed is observed for the various pulse durations.
5.3.5 Energy gain and conversion efficiencies

This section is assigned to the presentation of selected shots exhibiting high energy gain and discuss respective conversion efficiencies.

It is argued that the pulse energies given in this chapter are underestimating the actual output values (especially in the case of the negative gradient). Stimulated backscattering is exhibited in a much broader cone compared to the solid angle of seed or pump pulse. The seed extraction setup was
modified to collect the maximum amount of the backscattered signal (see Fig. 5.2). However, a gap of around 1.5 cm between the squared mirrors had to be retained because of the incoming pump pulse in the ~30 mrad off-axis geometry. Since the seed pulse and therefore the backscattering center is located at the edge of the mirror close to the incoming pump beam, a significant part of the backscattering hits the gap and is not acquired by the diagnostics. An distinct example is shown in Figure 5.27 where an amplified Raman signal as acquired by the spectrograph is depicted. A crossed periscope in front of the spectrograph flips the horizontal and vertical plane. Subsequently, the two squared mirrors are spatially resolved by the spectrograph. Mirror 1 is on the right side (in the seed direction) and collects the full profile of the unpumped seed pulse. Mirror 2 only collects a signal for the widened backscattering in case of spontaneous pump or amplification shots. The dashed yellow line indicates the area where the backscattering signal is lost for the diagnostics. Nearfield images show that the 2” squared mirror hit by the seed pulse is fully illuminated by the SRBS signal so that losses are not only expected in the gap between the mirrors. In order to proof that the signal on mirror 2 is not accounted for by spontaneous pump backscattering Figure 5.28 compares the Raman signal that is collected by respective mirror for a stimulated (blue) and spontaneous (green) backscattering pulse. For comparability the spontaneous backscattering shot with the highest signal count on mirror 2, which at the same time has the highest energy output, is selected. The amplified seed shot corresponds to the record shot of the same measurement series in terms of output energy, though amplification shots with even higher signal on the 2nd mirror can be found.

It is clearly shown that backscattering alone does not account for the signal collected by mirror 2.
Subsequently, it is proven that SRBS pulses exhibit wider solid angles and a significant part of the amplified pulse is not acquired by the diagnostics. Wide-angle scattering can occur in the kinetic regime $k_p \lambda_D \geq 0.29$ (which is reached for attainable electron temperatures of ~100 eV) when particle trapping induces transversal modulations of the EPW [102, 104, 109].

Let us now take a look at the high gain shots with the negative density gradient (peak plasma density $n_e \approx 1.6 \cdot 10^{19}$ cm$^{-3}$ (0.009$n_c$)). Amplified spectra of the two shots with highest energy output are compared to the transmitted seed spectrum (w/o plasma) and three spontaneous backscattering spectra with maximum energy output (Fig. 5.29). Unfortunately, the record amplification shot exceeded the measurement range of the pyroelectric energy sensor at that time. It is only possible to give a lower limit of the total output energy which is ≥975 µJ. In order to determine the energy transfer and efficiency the seed energy transmitted through the ionized target (135 µJ) and the peak energy of the spontaneous backscattering (140 µJ) have to be taken into account which results in a total energy transfer of 700 µJ and efficiency of 0.4%. This is the most conservative estimate since the lowest limit of the amplified pulse energy is used and losses originating from the wide angle SRBS signal not hitting the extraction mirrors are not considered. The spectrograph image shown in Fig. 5.28 actually depicts the Raman signal of aforementioned shot and indicates that indeed an intense part of the amplified seed profile is lost.

The spectra for the conical nozzle target (plasma density $n_e \approx 2.2 \cdot 10^{19}$ cm$^{-3}$ (0.013$n_c$)) are shown in Figure 5.30. Maximum output energy is 1660 µJ, deducting peak spontaneous backscattering and seed transmission energy leaves ~1315 µJ transferred from pump to seed. Conversion efficiency is therefore ~0.8%, again not considering losses arising from the incomplete SRBS pulse profile extraction. Though it should be noted that for the flat-top target SRBS backscattering angles are seen to be much smaller than for the gradient target. Therefore, losses should be much less significant compared to the negative gradient target. From a rough approximation (Gaussian seed profile interpolation) actual energy gain in the gradient profile is estimated to be at least comparable to the flat-top target.

However, direct nozzle comparison is not appropriate since for the trapezoid nozzle the target and therefore plasma length is doubled. As the main pulse of the pump has a comparably short pulse duration both pulses interact for ~1.4 mm. When only taking into account the FWHM of the pump pulse (3.6 ps) the optimized interaction length is ~0.5 mm $\left(\sim \frac{\tau_p c}{2}\right)$. Additional plasma length worsens the effects of ionization scattering, inverse Bremsstahlung and competing instabilities.
5.4 Summary

This chapter has presented the results of ultrashort, high bandwidth seed pulse amplification by SRBS in well-defined flat-top and gradient plasma density profiles. Direct comparison of opposite
Stimulated Raman backscattering in tailored plasma density gradients

sign gradient profiles, achieved by rotation of a 3 mm trapezoid nozzle, showed a strong mitigation of SRBS as well as spontaneous pump backscattering for the positive gradient (~15% higher detuning growth rate).

Results for all targets, including a flat-top profile generated by a 1.5 mm conical nozzle, indicate that the stimulated backscattering instability can be optimized for higher plasma densities and therefore increased pump-to-plasma frequency ratios ($\frac{\omega_n}{\omega_{pe}} \approx 10$) than the previously suggested range of $\frac{\omega_n}{\omega_{pe}} = 14 \ldots 20$.

In relation to the flat-top profile, output spectra for the negative gradient exhibit a superior bandwidth and increased redshift of the Raman peak at comparable plasma densities and relative pulse delays. It is suggested that efficiencies are similar but the SRBS signals originating from the ramp target show significantly increased solid angles which may be explained by particle trapping induced Langmuir wavefront bowing.

One-dimensional PIC simulations for the negative gradient profile are in good agreement with experimentally acquired spectral data for similar pulse and plasma parameters. The duration of the leading pulse of the SRBS pulse train as given by the simulations is ~75 fs. 90% of the pulse energy is contained in the first spike which is of great importance for the subsequent usage of parametrically amplified output pulses in future applications.
Chapter 6

High power seed pulse generation for stimulated Raman backscattering experiments

Stimulated Raman backscattering experiments in plasmas require a high energy pump pulse and a short seed pulse. The latter should be shifted to lower frequencies in regard to the pump pulse. Phase matching condition is fulfilled when the difference between the pump and seed frequency corresponds to the frequency of the plasma oscillations \( \omega_0 - \omega_1 = \omega_p \). Typical values for electron plasma densities are \( 10^{18} - 10^{19} \, \text{cm}^{-3} \) in order to trigger the Raman backscatter instability \( (n_e < \frac{1}{4} n_c) \). Therefore plasma frequencies are \( 5 \cdot 10^{13} - 10^{14} \, \text{s}^{-1} \) resulting in shifts of 50 to several hundreds of nanometer depending on the SRBS pump laser wavelength. PIC simulations showed that phase matching is not the only restriction to the frequency difference of the two pulses but in order to efficiently drive the SRBS instability \( \frac{\omega_o}{\omega_{pe}} \) should be between 14 and 20 [41]. This gives a tighter restriction on the shift of the seed pulse resulting in a frequency shift of 40 – 60 nm for Ti:sapphire and 50 – 100 nm for ND:glass systems. There are several methods to shift the laser frequency as discussed in chapter 3. A common method is utilizing Raman crystals where conversion efficiencies reach values up to 30\% for the first Stokes shift [42]. But input short pulse energies are usually below 5 mJ and cannot be upscaled due to damage thresholds (~0.1 – 2 GWcm\(^{-2}\)). Extending the focal region can keep intensities below damage threshold, but typical Raman crystals
lengths are only up to several centimeters and the (for solids) more distinct group velocity mismatch restricts phase matching lengths. Long gas cells are comparably cheap, easy to set up and allow meter scale gain lengths as well as the usage of different gases for variable frequency shifts whereas Raman crystals have fixed Raman shifting values. Aim of the experiments is to show the applicability of frequency shifting by Bessel beam pumping in gas cells for the seed generation in future SRBS experiments. Output powers should be \( P \geq 10 \text{ GW} \) in order to reach focal intensities of \( 10^{14} - 10^{15} \text{ W cm}^{-2} \) which are required seed pulse intensities for efficient SRBS [41]. Considering aforementioned restrictions due to SRBS plasma densities and pump-to-plasma-frequency ratios, at ND:glass systems (~1055 nm) shifted output pulses should have central wavelengths around 1100 – 1150 nm, whereas at Ti:sapphire system (~800 nm) it is opted for \( \lambda_c = 840 – 860 \text{ nm} \).

6.1 Bessel beam pumping setup

A general scheme for the setup is shown in Fig. 6.1. The pulses coming from the amplifiers of the respective CPA scheme laser system are sent to the compressors that are used to adjust the chirp (resp. pulse duration) for different regimes of nonlinear frequency conversion.

The pump pulses are then demagnified or magnified, depending on the required energy density. An aperture is placed before the axicon to adjust beam diameters and consequently caustic lengths. The use of diverging beams in order to modify the longitudinal intensity distribution is also possible but leads to non-constant focal spot sizes along the optical axis [43]. The axicon, placed behind the first aperture, produces quasi-Bessel beams propagating on the optical axis (chapt. 3.2.2). The 1” or 2” fused silica axicons are placed shortly before the gas cell to avoid nonlinear effects at the entrance window. The gas cell length must be chosen accordingly to the caustic length of the axicon (Eq. 3.6) to optimize gain and reduce intensity at the exit window.

Aiming for long gain lengths, small base angle axicons (\( \alpha = 0.25^\circ, 0.5^\circ, 1^\circ \)) are utilized producing caustic lengths up to a few meters and therefore equally require meter scale gas cells. Proper alignment of small angle axicons is crucial and can be done by utilizing the back reflection or generating a symmetric, homogeneous ring shape in the far field. Since alignment is very sensitive micrometer screws for the three spatial directions and the tip-/tilt angles are advantageous. The second aperture is placed at the exit of the gas cell, blocking the output beams not propagating on the optical axis (fundamental and anti-Stokes mode). From there the slowly diverging Stokes mode
can be collimated and sent to the different diagnostics, including single-shot autocorrelators, pyroelectric energy sensors, CCD cameras and fiber spectrometers. Working gases that are utilized during the experiments include sulfur hexafluoride (SF$_6$), nitrogen (N$_2$), isobutane (C$_4$H$_{10}$), methane (CH$_4$) and argon (Ar) with applied pressures that range from 100 mbar to 2 bar.

![Figure 6.1: Scheme for nonlinear frequency conversion by Bessel beam pumping in gas cells.](image)

6.2 Continuous redshifting in air, N$_2$, O$_2$ and SF$_6$ at the Jeti40 laser (Ti:sa)

The possibility of seed pulse generation for SRBS experiment utilizing few millijoule, $\tau = 10^{-14} - 10^{-12}$ s pulses for extended caustic pumping of various gases is presented. Continuous frequency shifts, supercontinuum generation and or shifting by stimulated Raman scattering (bound to the vibrational states in molecules) is observed. This section is differentiated to chapter 6.3 by the input pump pulse parameters. Energies are around one order of magnitude lower while the bandwidth is broadened by the same amount. Pulse durations tend to be smaller in order to achieve the required intensities on the optical axis.
6.2.1 Pump pulse parameters and experimental setup

The setup follows the general scheme shown in Fig. 6.1. Beam diameter before axicon is \( w_0 = 10 \text{ mm} \). Pump pulse energy is varied by adjusting amplifier timings and covers the range \( E = 2 – 20 \text{ mJ} \). The pulses can be either negatively or positively chirped by the grating compressor to obtain pulse durations \( \tau = 0.03 – 3 \text{ ps} \). Central wavelength and bandwidth is \( \lambda_c \approx 800 \text{ nm}, \) respectively \( \Delta \lambda \approx 60 \text{ nm} \) (spectra shown in chapt. 5, Fig. 5.7). The 1" fused silica axicon has a base angle \( \alpha = 0.25^\circ \). The gas cell is a 3 m acrylic glass tube with 5 mm fused silica windows under Brewster angle connected by ISO-KF40 flanges. Pressure inside the cell is controlled by a Bronckhorst flow controller. Investigated working gases are sulfur hexafluoride (SF\(_6\)), methane (CH\(_4\)), nitrogen (N\(_2\)), oxygen (O\(_2\)) and air. The pulse propagating on the optical axis is cut out by an aperture behind the gas cell and sent to an uncoated wedge redirecting the beam to the different diagnostics. The diagnostics include a fiber spectrometer with a silica based CCD array (OceanOptics USB2000), a pyroelectric energy sensor (Coherent EnergyMax) and a CCD camera to capture beam profiles. Intensity distributions along the pulse propagation axis (after Eq. 3.7) for typical laser parameters are shown in Fig. 6.2. It is important to notice that the maximum intensity scales inversely linear to the pulse duration, so that for shortest pulse duration \( \tau = 30 \text{ fs} \) maximum intensities are 28 times higher.

![Figure 6.2: Radial Bessel beam intensity distribution along the caustic for an axicon base angle 0.25°, 20 mm beam diameter, 850 fs pump pulse duration, 11 mJ (left) and 18 mJ (right) pump energy.](image-url)
6.2 Continuous redshifting in air, N\textsubscript{2}, O\textsubscript{2} and SF\textsubscript{6} at the Jeti40 laser (Ti:sa)

6.2.2 Experimental results

At first the impact of the pulse duration and chirp sign on the energy and spectrum of the axial mode is investigated. Experimental data of the chirp scans in sulfur hexafluoride (SF\textsubscript{6}) and methane (CH\textsubscript{4}) are shown in Fig. 6.3. The first vibrational mode of SF\textsubscript{6} has the spectroscopic wavenumber $\tilde{\nu}_1 = 774$ cm\textsuperscript{-1} \cite{44} which corresponds to the centrosymmetric stretch of the fluoride atoms. For methane the wavenumber for the first vibrational mode is $\tilde{\nu}_1 = 2917$ cm\textsuperscript{-1} \cite{45} corresponding to a centrosymmetric stretch of the hydrogen atoms. Subsequently, the frequency shifted spectrum of a 800 nm pulse can be calculated which yields spectral peaks at 853 nm ($\tilde{\nu}_1$(SF\textsubscript{6})), respectively 1043 nm ($\tilde{\nu}_1$(CH\textsubscript{4})). The vibrational dephasing times are $T_2 = 6$ ps (SF\textsubscript{6}) \cite{46} and $T_2 = 16$ ps (CH\textsubscript{4}) \cite{47}. Since pump pulse durations are generally $T_1 < \tau < T_2$, Raman interactions are expected to take place in the transient regime.

For negatively chirped pulses in SF\textsubscript{6} a stable peak at around 855 nm (which fits perfectly when considering the precise central wavelength of the pump pulse $\lambda_c = 802$ nm) develops for pulse durations $\tau > 0.5$ ps which is likely coupled to the first vibrational mode. For positive chirps equal spectral components at the expected wavelength window appear, but for both directions the majority of spectrum and therefore intensity is found at lower or higher frequencies.

In CH\textsubscript{4}, vibrational Raman sidebands appear but the central wavelength is shifted to lower (neg. chirp) or higher (pos. chirp) frequencies presumably due to spectral phase dependent SPM and group velocity mismatch determined SRS phase matching (Fig. 6.4). However, frequencies are too low to be appropriate seeding pulses for SRBS at Ti:sapphire lasers. The majority of the spectral intensity is found around the initial wavelength accounted for by supercontinuum generation.

![Figure 6.3: Chirp depended output spectra for sulfur hexafluoride and methane. White dashed lines indicate the central wavelength of the pump spectrum.](image-url)
The results for air, diatomic nitrogen and oxygen are shown in Fig. 6.5. The Raman shift of the first vibrational mode for N$_2$ and O$_2$ is 2331 cm$^{-1}$, respectively 1555 cm$^{-1}$ [45]. Spectral peaks are expected at 983 nm (N$_2$) or 914 nm (O$_2$). The data for air reproduces a weighted overlap of the diatomic nitrogen and oxygen spectra. No spectral components related to pure vibrational Raman scattering are found which may be explained by comparably low Raman scattering cross-sections for O$_2$ and N$_2$, that are almost one order of magnitude lower compared to methane [54]. All gases (methane excluded) exhibit a chirp dependent continuous redshift of a comparably narrow bandwidth peak. Continuous self-frequency shifting was first observed in optical fibers and waveguides and described as an interplay between SPM and Raman scattering [48]. [49] suggests impulsive excitation of molecular vibrations, but here the effect occurs for $\tau >> T_1$, that is when pump pulse durations are much longer than the vibrational period, where e.g. $T_1 = 43$ fs for $\tilde{v}_1$(SF$_6$).

The data suggests that for methane and sulfur hexafluoride pure vibrational Raman components...
emerge but the dominating effect of the continuous frequency shifting must be assigned to other processes. Excitations of electronic and rotational coherences, changing the refractive index for the trailing and leading edge (due to inertial responses) and therefore leading to a SPM induced redshift is suggested by several publications [50, 51, 52].

Measurements of axial pulse energies for \( \text{N}_2 \) and \( \text{SF}_6 \) (corresponding to the data in Figs. 6.3, 6.5) are shown in Fig. 6.6. At first a gradual increase in temporal chirp increases the energy of the axial beam which then reaches a maximum at a specific pulse duration before finally declining. The data is consistent to simulations for similar laser parameters by Bessel beam propagation in air [53].

Maximum conversion efficiency is 6% (9%) for a shift of the central wavelength to 830 nm (860 nm), obtained in \( \text{N}_2 \) (\( \text{SF}_6 \)). Maximum redshifts do not necessarily coincide with highest conversion efficiencies.

Optimum pump chirp values for high output energies are used for the subsequent measurement of the output spectra as a function of the pump energy (Fig. 6.7). In case of nitrogen increasing pump energies lead to continuously higher frequency shifts as in the case of decreasing pulse duration, indicating that the frequency shift is determined by pump pulse intensity. After a certain threshold

![Figure 6.6: Chirp dependent output energy in \( \text{N}_2 \) (left) and \( \text{SF}_6 \) (right).](image)

![Figure 6.7: Output spectra as a function of pump energy at two different pressures in \( \text{SF}_6 \) and 1 bar nitrogen.](image)
the strongly redshifted component starts to disappear, coincident with decreasing output energies and supercontinuum generation. Decreasing energies are caused by stronger ionization of the working gas, leading to diffraction and scattering losses of the pump pulse. Aforementioned cases exhibit a break-up of the beam profile into multi-mode structures. The SF$_6$ spectra equally reproduce the data from the corresponding chirp scan. Especially on the negative chirp side little or no continuous frequency shift is measured, again suggesting that the frequency shift is rather determined by vibrational Raman scattering than SPM. Intensity thresholds for the onset of Raman scattering are dependent on the working gas pressure and here calculated as being around $1 \cdot 10^{12}$ Wcm$^{-2}$ (Fig. 6.2). The pressure scan in N$_2$ shows that equally to varying pump intensity, continuous shifting can be achieved by changing gas cell pressures (Fig. 6.8). A threshold is found ($\geq 1500$ mbar) where plasma generation and deteriorating beam profiles starts to diminish the shifted pulse. In order to obtain seed pulses for SRBS experiments it is also possible to utilize the setup for supercontinuum generation. If the spectrum is sufficiently broadband there must be spectral components satisfying phase matching condition for SRBS. High intensity induced supercontinuum spectra for three different gases are shown in Fig. 6.9. Best results in terms of broadband pulse generation are obtained in methane, where the spectrum stretches far to the near infrared (NIR) region. Quantum efficiency of the Si-detector is very low in that region and cuts off at 1100 nm so that the NIR spectrum is expected to be more pronounced and likely stretches further than 1100 nm. Conversion efficiencies are around 10% with the highest value (14%) for SF$_6$. Besides the ability to shift the frequency of the input pulses and having a spatial separation to the fundamental mode, Bessel beam pumping exhibits more features that make it viable not only in the case of SRBS seed generation. Those features include high beam qualities (below the multi-filamentation threshold) and short pulse durations. Fig. 6.10 shows a typical beam profile of the axial mode. $M^2$ is estimated as being below 1.5, theoretically a perfect Gaussian mode is expected after breakup of the filament caused by a lack of background reservoir intensity. An indirect pulse duration measurement, by comparing second harmonic generation efficiency to another laser system (similar spectrum after inserting 850 nm bandpass filter) yields 200 - 300 fs for the shifted pulse in SF$_6$ at pump pulse durations $\tau = 800$ fs.
6.2 Continuous redshifting in air, N\textsubscript{2}, O\textsubscript{2} and SF\textsubscript{6} at the Jeti40 laser (Ti:sa)

Figure 6.9: Supercontinuum spectra generated in CH\textsubscript{4}, N\textsubscript{2} and SF\textsubscript{6} for 18 mJ bandwidth limited pulses.

Figure 6.10: Beam profile of the axial output pulse (SF\textsubscript{6}).

6.2.3 Summary

It was shown that Bessel beam pumping in gas cells can generate the required frequency shifts to \( \approx 850 \text{ nm} \) for the working gases SF\textsubscript{6}, O\textsubscript{2} and N\textsubscript{2}. A parameter window to obtain short, redshifted pulses with a high quality beam profile is established. The central wavelength of the axial pulse can be continuously shifted by varying pump intensity or gas pressure. The tuning range comprises up to 60 nm. Best conversion efficiencies (up to 9\%) are achieved in sulfur hexafluoride, producing \( \sim 1.8 \text{ mJ} \) output energies with good shot-to-shot stability. Resulting peak powers are \( \sim 7 \text{ GW} \). Alternatively, superbroadband pulses can be generated with conversion efficiencies up to 14\%.

Although for the seed pulse generation at SRBS experiments continuously redshifting is proposed as the preferable method since seed pulse energies in the relevant wavelength window are expected to exceed those for supercontinuum generation. Additionally, better conversion efficiencies for white light generation can be achieved by using waveguides (e.g. gas-filled hollow-core fibers) as described in chapter 5.2.1.
6.3 Vibrational, multi-order SRS in SF₆ and isobutane at the PHelix laser (Nd:glass)

In the previous experiment frequency shifted pulses with energies up to 1.8 mJ were generated by Bessel beam pumping for input energies ~20 mJ and pulse durations ~1 ps. An equivalent experiment is set up in the X-ray laboratory at the Phelix laser facility. The Nd:glass system lases at a higher wavelength and exhibits a narrower bandwidth which increases the bandwidth limited pulse duration to ~500 fs. At the same time, around one order of magnitude higher pulse energies can be utilized resulting in on-axis intensities of the Bessel beam higher or similar to those in chapter 6.2.

When higher input energies can be translated to higher output energies, redshifted seed pulses may be able to directly enter the pump pulse depletion regime in SRBS experiments. When output pulse durations are sufficiently short, respectively when the bandwidth supports compression down to the timescales of a plasma period, seed pulses can theoretically mitigate the decreased SRBS efficiency when operating in the strong wavebreaking regime [55].

Ideally, input pulse durations for the Bessel beam pumping should be appropriate for SRBS pumping at the same time. Pump pulse durations in SRBS experiments depend on target length and focal intensity limits, typical values are a few to several tens of picoseconds. Having the possibility to operate with the same pulse duration for the seed generation in the gas cell and SRBS pumping simplifies the experimental setup since no additional compressor is needed.

6.3.1 Pump pulse parameters and experimental setup

The pulse coming from the compressor in the X-ray laboratory is demagnified by a telescope and sent on a 2" fused silica axicon with base angle \( \alpha = 0.5^\circ \) or \( 1^\circ \). Typical beam diameter is 18 mm. Intensity distributions are shown in Fig. 6.11. The 2.8 m gas cell consists of several pieces of vacuum tubes connected by CF63 flanges. The entrance and exit CaF₂-windows are 6 mm thick and anti-reflective coated. CaF₂ was chosen because it offers a low dispersion and high damage thresholds. Working gases are isobutane (C₄H₁₀), sulfur hexafluoride (SF₆) and argon (Ar). Gas
6.3 Vibrational, multi-order SRS in SF$_6$ and isobutane at the PHELIX laser (Nd:glass)

Pressure is adjusted by a Bronckhorst flow controller. The frequency shift of the generated seed pulses should be between 500 and 800 cm$^{-1}$ considering SRBS phase matching at typical plasma densities ($\omega_0 = \omega_1 + \omega_p$) and suggested pump-to-plasma-frequency ratios ($\frac{\omega_0}{\omega_p} = 14 \ldots 20$). Gaseous SF$_6$ offers a wavenumber shift of 774 cm$^{-1}$ for the first vibrational mode ($\tilde{\nu}_1$). Gaseous isobutane has a Raman shift $\tilde{\nu}_1 = 2880$ cm$^{-1}$ but also offers a shift in the required range for a higher vibrational mode $\tilde{\nu}_7 = 794$ cm$^{-1}$ at a comparably high relative Raman scattering cross section [45].

It should be noted that vibrations from the (single bonded) functional groups C-I, C-Br, C-Cl, C-S and O-O show strong Raman scattering in the desired range but it is difficult to find those groups in (non-hazardous) molecular gases at room temperature. The generated axial beam is separated from the fundamental and anti-Stokes modes by an aperture behind the gas cell. Our measurements include spectral and energy dependence of the axial Stokes beam on pump chirp, input energy and beam diameter. Pump energies are $E = 100 \ldots 300$ mJ with pulse durations of 500 fs (bandwidth limit) to several picoseconds. Pump pulses are merely positively chirped since the compressor setup does not support overcompensating chirp introduced by the stretcher. The central wavelength and bandwidth of the Nd:glass based system is $\lambda_c = 1055$ nm respectively $\Delta \lambda = 6$ nm, measured by an Ocean Optics HR4000 fiber spectrometer (Fig. 5.11). Behind the gas cell, generated pulses are collimated and sent to the different diagnostics by pellicle beamsplitters. In order to record redshifted spectra in the near-infrared region an Ocean Optics NIRQuest512 with an indium gallium arsenide (InGaAs) array detector is used giving access to the wavelength range of 900 – 2100 nm. The fiber spectrometer is connected to an integrating sphere ensuring spectral acquisition of the complete beam profile. The beam profile diagnostic utilizes second harmonic generation in a KDP crystal and subsequent detection with a Si-CCD camera. Pulse duration is measured by a single-shot autocorrelator and energy by a pyroelectric energy sensor.

Figure 6.11: Radial Bessel beam intensity distributions along the caustic for 18 mm beam diameter, 300 mJ pump pulse energy at $\alpha = 0.5^\circ$, $\tau = 4$ ps (left) and $\alpha = 1^\circ$, $\tau = 8$ ps (right).
6.3.2 Experimental results

The recorded spectra show a different behavior than those acquired at the Jeti40. No continuous frequency shifts are observed. For pump pulse durations $\tau \geq 1$ ps output pulses exhibit a constant spectral behavior directly coupled to the first vibrational mode of the corresponding gas molecules. Therefore output wavelengths of the Stokes pulses can be calculated with

$$\lambda_S = \frac{\lambda}{1 - \frac{\lambda}{\nu \cdot [m^{-1}]}}$$

which yields $\lambda_S = 1149$ nm ($\nu_1(SF_6)$) and $\lambda_S = 1515$ nm ($\nu_1(C_4H_{10})$). Argon is chosen to confirm that frequency shifts are due to vibrational Stokes scattering which cannot occur in atomic gases.

Figure 6.12 shows spectra of the Stokes mode for the three different gases. Argon does not display any significant shift of the central wavelength which holds true for all scans conducted ($E = 200 – 300$ mJ, $\tau = 0.5 – 12$ ps). The spectral shifts for SF$_6$ and isobutane coincide with the expected values from literature for the first vibrational Raman mode. It can be seen that this Raman mode is outgrowing all other potential vibrational modes which have lower Raman cross-sections. In the case of SF$_6$ and isobutane residual spectra of the fundamental wavelength (1055 nm) are due to imperfections of the axicon tip or aperture placement before complete separation of the k-vectors [56]. The isobutane peak at $\sim 1.5$ $\mu$m exhibits a strong SPM broadened base due to higher intensities compared to the Stokes pulse generated in SF$_6$.

Figure 6.12: Raman Stokes shift in sulfur hexafluoride and isobutane.
6.3 Vibrational, multi-order SRS in SF$_6$ and isobutane at the PHELIX laser (Nd:glass)

Figure 6.13 shows chirp scans in SF$_6$ for two different axicon base angles ($\alpha = 0.5^\circ$, 1°) and isobutane ($\alpha = 1^\circ$). For bandwidth limited pump pulses, high axial intensities generate a supercontinuum with redshifted central wavelength (see also blue curve in Fig. 5.15). Increasing the pump chirp leads to an onset of intense Stokes pulse propagation on the optical axis. The intensity of the first order Stokes pulse can be sufficient to equally act as a pump pulse. In case of SF$_6$ this leads to multiple peaks at higher wavelengths. Aforementioned spectral characteristics are exclusively higher orders of the $\nu_1$ vibrational mode of SF$_6$, whereas the second order (1260 nm) is generated by Stokes scattering from the first order (1149 nm) and so forth. A further increase in pump chirp leads to the extinction of those higher orders since intensity of the Stokes mode drops below the value sufficient to act as a pump itself. A maximum energy of 8 mJ is measured for the first Stokes mode in SF$_6$ which yields an efficiency of 3% for converting 1055 nm into a spatially separated 1149 nm pulse.

Isobutane is expected to equally generate higher orders but already the second order Stokes radiation (2688 nm) is well outside the detection range of the fiber spectrometer. A maximum energy of 30 mJ is measured behind the gas cell. However it is unclear what percentage accounted for by the first order since pyroelectric energy sensors have a wavelength range up to tens of microns and no appropriate shortpass filter was available.

To understand the effects of different pump pulse durations and axicon angles in terms of phase matching / temporal overlap of the pump and generated Stokes pulse, it is necessary to take a look at the group velocity mismatch (GVM). The GVM is defined as the difference of the inverse group velocities

$$GVM = \frac{1}{v_{g,B}} - \frac{1}{v_{g,S}}$$
where $\nu_{g,S}$ is the group velocity of the Stokes pulse and $\nu_{g,B}$ the group velocity of the pump Bessel pulse given by

$$\nu_{g,B} = \frac{c/n_G}{\cos \theta \cdot (1 - (\omega \cdot \frac{\partial \theta}{\partial \omega}) \cdot \tan \theta)}$$

with $n_G$ refractive index of the working gas, $\omega$ angular frequency and $\theta = \alpha \cdot \left(\frac{n_a}{n_{air}} - 1\right)$ the cone angle of the Bessel beam in the small-angle approximation. An estimation for the refractive index of sulfur hexafluoride can be made by [57]

$$n = A \cdot \left(\frac{P_G}{RT}\right) + B(T) \cdot \left(\frac{P_G}{RT}\right)^2$$

where $P_G$ is gas pressure in atm, $R$ gas constant, $T$ temperature and $A$, $B(T)$ are coefficients that need to be determined experimentally. A calculation with the corresponding values ($p = 0.89$ atm, $T = 294$ K, $A = 17.5$ and $B = 7602$) yields $n_{SF6} = 1.0076$. Subsequently the GVM is approximately 600 fs/m ($\alpha = 1^\circ$) and 150 fs/m ($\alpha = 0.5^\circ$). For the 0.5° axicon this should be negligible, for the 1° axicon the temporal walk-off over the 2.8 m long gas cell has to be considered as a restriction to the gain length. Unfortunately no conclusive study about the sign of the dispersion in the relevant wavelength range can be found. Though results in [58] indicate an anomalous dispersion which would induce an even earlier mismatch between the pulses considering that Stokes pulses are delayed in respect to the pump pulse for transient SRS [59]. Either way, the increase of Stokes intensity for longer $\tau$ as seen in Fig 6.13 is explained by elongated phase matching of the two pulses. For instance at 0.9 ps (yellow curve in Fig. 6.14) a minor spectral broadening is observed, indicating that pump intensity is close to a value avoiding competing nonlinear effects. However Stokes pulse intensity still increases up to 2.8 ps due to an extended overlap overcompensating declining pump intensities. Identical measurements show for the 0.5° axicon an increase of the first order Stokes mode only as long as broadband generation is declining.

Looking at the effect of two different axicon base angles in Fig. 6.13, it is obvious that the longer caustic length is not able to compensate for the drop of the on-axis pump intensity by a factor of 5, when switching from $\alpha = 1^\circ$ to 0.5° (for $\tau = \text{const.}$). For the 0.5° axicon the first Stokes mode starts to diminish sooner and therefore higher orders start to decrease earlier. The residual pump spectrum is more pronounced, indicating that the caustic length stretches beyond our separating aperture behind the gas cell and weak white light generation is observed in the exit window of the
gas cell. The corresponding caustic lengths and Bessel pump intensities for both axicons at pump chirps suppressing higher order Stokes generation are shown in the previous chapter (Fig. 6.11). As follows, on-axis pump intensity should not exceed \(8 \cdot 9 \times 10^{12} \text{ Wcm}^{-2}\) if extraction of a single Raman mode is required. In order to avoid SPM and extract (unmodulated) multi-order Stokes pulses, on-axis pump intensity must not exceed \(I = 4 \cdot 5 \times 10^{13} \text{ Wcm}^{-2}\).

Figure 6.15 shows that an increase in beam diameter prior to Bessel beam focusing leads to an increase of first Stokes intensity and subsequently higher Stokes orders. It is important to note that the beam diameter was adjusted by the aperture before the 0.5° axicon. Here intensity of the collimated pump is approximately constant over the whole diameter (top-hat beam). As a consequence a larger diameter increases input energy by a factor \(\left(\frac{r_2}{r_1}\right)^2\) with \(r_2 \geq r_1\). Therefore, similar to the case of switching to smaller base angles axicons the caustic length is prolonged but not at an expense of Bessel beam intensity. For maximum beam diameter we are able to generate up to fifth order Stokes pulses at 1780 nm. For the high harmonic Stokes generation second order intensity can be equal to the first order giving the possibility to use higher orders at comparable efficiencies if lower frequency output pulses are required. Stokes generation can equally be modified by adjusting working gas pressures (Fig. 6.16). Since the Raman gain coefficient scales linearly with the population density for the Raman transitions (Eq. 3.4), raising gas pressures lead to an increase in Raman scattering. This can be of particular interest if laser parameters are fixed, for instance in SRBS experiments where laser pulse duration and energy is adapted to SRBS pumping requirements.
Pulse duration measurements of the first Stokes mode by a single-shot autocorrelator indicates $\tau_S \approx 300$ fs for an input pump pulse duration of 2 picoseconds (Fig 6.17). Subsequently maximum output power for the first Stokes mode in SF$_6$ is 27 GW. The shortest possible pulse duration supported by the measured bandwidth of the first Stokes mode can be calculated with

$$\Delta\tau = \frac{\lambda^2}{\Delta\lambda \cdot c} \cdot tbp$$

where $\lambda_S = 1149$ nm, $\Delta\lambda_S = 30$ nm and $tbp$ (time bandwidth product) = 0.315 for sech$^2$-shaped pulses, which yields $\Delta\tau_S = 46$ fs. Implementing a chirped mirror or additional compressor setup for the generated pulse could therefore further increase the output power by a factor of 6.

A typical beam profile of the first order Stokes mode is shown in Fig. 6.18, exhibiting a nearly perfect Gaussian profile with a focal spot diameter of 100 $\mu$m. The beam profile was measured after second harmonic generation (SHG) since quantum efficiency of the silica CCD approaches zero for wavelengths $\lambda \geq 1100$ nm. The effect of the SHG is removed for the far field image and subsequent focus diameter.

Figure 6.16: Higher order Stokes generation for various gas-cell pressures (SF$_6$).

Figure 6.17: Autocorrelation measurement of the first order Stokes mode in SF$_6$ at an input pump pulse duration of 2 ps.

Figure 6.18: Typical beam profile of the axial (first-order Stokes) output pulse in SF$_6$. 
6.3 Vibrational, multi-order SRS in SF₆ and isobutane at the PHELIX laser (Nd:glass)

6.3.3 Summary

A new method for generating multi-millijoule seed pulses at stimulated Raman backscattering experiments is proposed. The setup utilizes Bessel beam pumping in Raman active gases where the wavelength shift is determined by the first vibrational mode of the gas molecule or its higher harmonics. A parameter range for efficiently driving the SRS process depending on gas pressure, gain length and intensity is shown. Intensity of the first order Stokes pulse is restricted by the onset of higher harmonic generation. Frequency shifted output pulses are short (3 digit fs range), spatially separated from the fundamental and anti-Stokes modes, have a high beam quality and comparably high energies. In [60] Bessel beam propagation in a 9 cm Raman crystal yielded 3.2 mJ for the Stokes pulse, however input pulse duration was $\tau = 15$ ns to stay below the damage threshold of the Raman crystal. Pulse duration was not measured, assuming one order of magnitude pulse compression as was shown for Stokes generation in Raman crystals [61], yet the proposed setup in this work generates redshifted pulses with higher energies and 4 - 5 orders of magnitude higher output powers. A direct comparison to the seed generation in similar experimental works covering SRBS shows seed powers of 13 MW (2 µJ, 150 fs) for (a more sophisticated) optical parametric amplifier setup [62], 16 MW (8 µJ, 500 fs) for SRS in a boron nitride crystal [12] and 2.5 GW (500 µJ, 200 fs) for SPM under filamentation in a gas cell [63]. Whereas all aforementioned cases require an additional compressor setup.

Results presented in this chapter show that high power seed pulses at phase matched frequencies for SRBS can be generated via Bessel beam pumping in SF₆. Input pump pulse durations can at the same time be viable for SRBS pumping, subsequently removing the necessity of an additional compressor or chirped mirror setup. However, if an appropriate group delay dispersion is introduced, bandwidths suggest that pulse duration as short as tens of femtoseconds can be acquired.

To decrease the setup length, usage of a phase matched SRS seeding pulse is proposed since higher efficiencies can be reached over equal gain lengths [64, 65].
6 High power seed pulse generation for stimulated Raman backscattering experiments
Chapter 7

Summary & outlook

This thesis presents results for plasma-based optical parametric backscatter amplifiers at Ti:sapphire laser systems and the generation of multi-millijoule seed pulses for stimulated Raman backscattering experiments.

At the Gemini laser amplification of $\sim10^{14}$ W cm$^{-2}$ seed pulses by multi-joule $\sim10^{16}$ W cm$^{-2}$ pump pulses to energies exceeding the current record for plasma-based optical parametric amplifiers by a factor of $\sim1.35$ are obtained [92]. Output powers surpass previous results by almost one order of magnitude while short seed pulse durations and or comparably low electron plasma densities ($\sim0.009n_e$) ensure a high quality spatial seed profile. The wavelength shift between seed and pump pulse is around 10 nm and should therefore only support efficient SRBS for plasma densities one to two orders of magnitude lower than utilized in the Gemini campaign. However, efficiencies up to $\sim2\%$ are achieved at ever-increasing plasma densities. Energy gain curves show high spectral energy gain at the central wavelength of the initial seed spectrum. Output spectra are robust to changes in plasma density or relative pulse delays which is in contrast to results of the stimulated Raman backscattering beamtime at Jeti40. Furthermore, one-dimensional PIC simulations with infinite ion masses were not able to reproduce seed gain spectra or pulse durations for aforementioned pulse and plasma parameters. Therefore, it is suggested that the corresponding parametric amplification process couples to a quasi-ion plasma wave in the so-called strongly coupled stimulated Brillouin backscattering regime which was first theoretically described in 2006 [91] followed by experimental evidence in recent years [40, 92, 93]. Sc-SBBS allows for amplified seed pulse durations smaller than ion acoustic timescales which makes the process a viable
alternative to SRBS or superradiant Raman amplification when it comes to the prospective
generation of ultra-high intensity laser pulses.
For the stimulated Raman backscattering beamtime at the Jeti40 laser ultrashort, broadband
$10^{14}$ Wcm$^{-2}$ seed pulses are generated by self-phase modulation in a argon-filled hollow-core fiber
with subsequent compression by chirped mirrors. The seed pulses are overlapped with $10^{15}$ Wcm$^{-2}$
pump pulses in well-defined flat-top and gradient density profiles. Previous measurements
addressing SRBS in plasma gradients were performed by step-ionization schemes or pulses
obliquely injected into expanded plasma channels [95, 99]. The generation of a neutral density and
subsequent plasma density gradient by a trapezoid nozzle allows for an extended, smooth gradient
profile which can be precisely characterized by tomographic interferometry. Subsequently, the
necessity of running simulations for different ionization steps, additional ionization pulses and or
the implementation of an interferometer into the experimental setup vanishes.
The data acquired for the positive and negative density gradient (achieved by a simple rotation of
the trapezoid nozzle) shows that SRBS efficiency strongly depends on the algebraic sign of the
gradient. Though the pump chirp rate is large (~60 nm @ 3.6 ps) and the plasma gradient not steep
enough to significantly reduce detuning, strong mitigation of spontaneous pump backscattering as
well as stimulated Raman backscattering for the density gradient increasing the detuning growth
rate $q$ in backscatter direction (by ~15% in relation to the opposite sign gradient) is found.
We have found that pump-to-plasma frequency ratios lower than the previously suggested range of
$\omega_p/\omega_{pe} = 14 \ldots 20$ optimize SRBS efficiencies for both the flat-top as well as the gradient target, while
seed profiles show no sign of filamentation [41].
Furthermore, we have shown that the negative plasma gradient (as seen by the positively chirped
pump) enables larger output bandwidths of the SRBS pulses and seems to shift the Raman peak to
lower frequencies at similar plasma densities as compared to the flat-top profile. Additionally,
SRBS cone angles are enlarged by the gradient profile which may indicate the emergence of strong
wavefront bowing (as described e.g. in [104, 109]). One-dimensional PIC simulations that are in
good agreement with the acquired spectral data show output pulse durations of 75 fs while the
leading spike contains ~90% of the energy, indicating SRBS in the strong wavebreaking regime
[110].
In the last part of this work, a new method for generating multi-millijoule seed pulses for
stimulated Raman backscattering experiments is proposed. The setup utilizes (axicon-) Bessel
beam pumping in Raman active gases where the wavelength shift is determined by the first
vibrational mode of the gas molecule or its higher harmonics. A parameter window for efficient
driving of transient stimulated (molecular) Raman scattering (SRS) is established. The easy-to-
implement setup allows for multiple orders of magnitude higher output powers compared to similar shifting methods that have been utilized in previous SRBS experiments [12, 60, 62, 63]. It is shown that frequency shifted output pulses are compressed to the three-digit fs range (at input pulse durations of several picoseconds), they are conveniently spatially separated from the fundamental and anti-Stokes modes and exhibit a high beam quality. The bandwidth of the output pulses (e.g. for the working gases sulfur hexafluoride and isobutane) support pulse durations down to a few tens of femtoseconds. It should be noted that the meter-scale setup may be shortened by the implementation of copropagating, phase-matched SRS seeding pulses injected on the optical axis as it was shown for Gaussian focusing in high pressure cells [64, 65].

Plasma-based optical parametric amplification is at the moment seen as the only possible option to surpass current intensity limits of \(~10^{22} \text{ Wcm}^{-2}\) by up to several orders of magnitude [110, 111]. It would also allow for much more compact and cost efficient laser systems to be set up since the final (high) intensities are generated in the target plasma which does not have a damage threshold. Therefore, in CPA based systems the diameters of beamline mirrors and gratings could be significantly decreased. Solely the optics handling the parametrically amplified seed pulses would require large diameters or the implementation of plasma mirrors. Ultrahigh intensities (\(\geq 10^{23} \text{ Wcm}^{-2}\)) will make it possible to enter new regimes (e.g. relativistic ion plasmas), experimentally verify present-day theory and improve or enable applications such as particle accelerators, \(\gamma\gamma\)-colliders, creation of superhot matter or fast ignition fusion [17, 18, 19, 21].

Two plasma-based parametric instabilities are viable when it comes to the generation of ultrahigh intensity laser pulses. Stimulated Raman backscattering and corresponding regimes (e.g. superradiant Raman amplification) mediated by an electron plasma wave or the strongly coupled stimulated Brillouin backscattering mediated by a quasi-ion wave. Latter has the advantage that almost same frequency seed pulses can be used, it is robust to plasma inhomogeneities (density or temperature), requires shorter interactions lengths, has a lower energy loss to the plasma wave (following Manley-Rowe relations) and supports higher plasma densities and pump pulse amplitudes than SRBS [33, 40, 90, 91, 93, 96]. The main advantage of SRBS is that it supports compressed output pulse durations that are almost one order of magnitude smaller compared to sc-SBBS. Therefore, it theoretically supports higher peak intensities and is also suggested to be superior in terms of amplification ratios [33, 37].

Although we were able to show that experimental results can be improved, experimentally acquired efficiencies (for both processes) are only up to a few percent and still far from the theoretical predictions for SRBS [11, 31, 41, 112, 113, 114] and sc-SBBS [86, 91, 96]. In recent years several
processes have been proposed as being responsible for the mitigation of energy transfer from pump to seed pulse. Including optical frequency chirps [24, 108], wavebreaking [27, 31, 32, 34], particle trapping [102, 117], Landau damping [28, 37, 112], plasma wave filamentation [115], premature or amplified seed Raman/Brillouin forward- and backward scattering [29, 33, 37, 41] and modulational instabilities [25, 26, 63, 116]. One promising method to reduce the negative effect of wavebreaking is to operate in the coherent wavebreaking (CWB) regime of Raman amplification which requires seed pulse durations shorter or comparable to a plasma period \( \tau_1 \leq \frac{1}{\omega_p} \) and sufficiently high seed intensities \( I_1 \geq 10^{16} \text{ Wcm}^{-2} \) [55]. For future SRBS experiments, such parameters should be accessible (w/o setting up a separate laser system) when generating frequency shifted seed pulses by Bessel beam pumping of Raman active gases as presented in this thesis. The author hopes that the conducted studies help to improve the setups of upcoming experiments and advance the understanding of plasma-based optical parametric amplification processes on the journey to next generation laser systems.
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