Analysis of radiation damage in heavy-ion irradiated ionic crystals by magnetic resonance*

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We explore and expand the capabilities of magnetic resonance techniques, mainly nuclear magnetic resonance (NMR) but also electron spin resonance (ESR), as a tool for the characterization of radiation damage in solids. At present, we are focussing on ionic crystals, mainly lithium fluoride (LiF), which offers two suitable probe nuclei for NMR. In contrast, ESR is directly susceptible to paramagnetic defects and conduction electrons e.g. in metallic colloids. In 2012 our research concentrated on three topics: (a) Utilizing spatially resolved field-cycling (FC) NMR data to obtain information about the distribution of ion-beam induced defects, (b) using NMR spectroscopy to detect the presence of fluorine gas within irradiated samples, and (c) examining defect dynamics above room temperature by ESR and spatially resolved NMR after a series of stepwise annealing experiments.

(a) Common NMR relaxation theory in the presence of paramagnetic centres in alkali halides predicts a strong B-field dependence of the relaxation rate, depending on the distribution of defects within the sample [1]. This effect is supported by our data recorded by FC NMR. However, so far literature provides only analytical and approximate solutions of the resulting differential equation for homogeneous distributions. We are currently working on a numerical approach to calculate relaxation rates for any given distribution. By comparing our model with data derived from samples with known distributions, we hope to deduce from FC NMR the topology of unknown defect structures.

(b) The formation of molecular fluorine gas formed in neutron-irradiated LiF was first observed by CW-NMR spectroscopy some five decades ago [2]. Until now it was just speculated that fluorine gas also forms in samples irradiated with heavy ions having a strong radial gradient of the dose deposited around the ion track. Recently, we identified for the first time F_2 molecules in several LiF crystals irradiated with 6×10^{11} to 2×10^{12} ions/cm² by NMR spectroscopy. We have evidence that the appearance of fluorine molecules requires an energy loss above 10 keV/nm where tracks consist of a heavily damaged core zone of few nm in diameter [3]. At present, the nature of this track core is not yet fully understood.

c) With CW-ESR spectroscopy we detected the formation of metallic Li colloids in LiF at temperatures above 600 K during annealing experiments. Li colloids in heavy-ion irradiated LiF samples were already identified by optical absorption spectroscopy [4]. However, ESR allows us to gain access to more heavily irradiated samples where the large color centers concentration leads to a high optical density preventing quantitative analysis of optical absorption spectra.

By spatially resolved NMR relaxation rate measurements on a stepwise annealed LiF crystal, we observed a remarkable drop of the relaxation rate below the level found in the unirradiated part of the crystal (Figure 1). This effect appeared at a temperature which corresponds to the recently observed ion-beam induced formation of impurity magnesium colloids [4]. We thus tentatively attribute this effect also to the formation of impurity clusters. The concentration of paramagnetic impurities is decisive for the relaxation rate in unirradiated crystals.

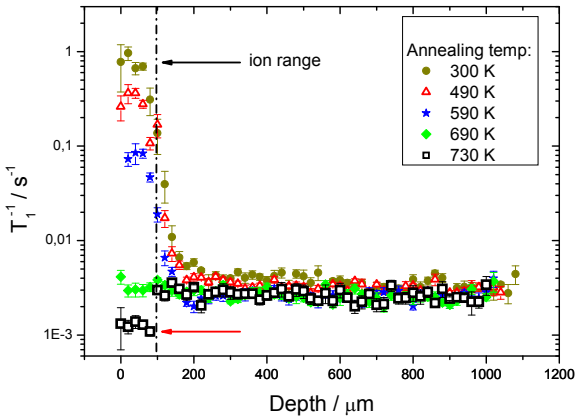

Figure 1: Depth profiles of nuclear spin-lattice relaxation rates recorded for a LiF crystal irradiated with 2.2-GeV ions of 6×10^{11} ions/cm². The sample was stepwise annealed for 20 min at different temperatures up to 730 K. Note the drop of relaxation rates at 730 K (red arrow) to a level below the value of the unirradiated, crystal (depth >100 μm).

References

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* Work supported by DFG, No. FU 308/12.
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