Ultrafast Magnetostriction of Antiferromagnetic Holmium studied by Femtosecond X-Ray Diffraction

Daniel Schick*, **Alexander von Reppert, Matthias Rössle**

Institut für Physik und Astronomie, Universität Potsdam, Karl-Liebknecht-Straße 24-25, 14476 Potsdam, Germany

Matias Bargheer

Institut für Physik und Astronomie, Universität Potsdam, Karl-Liebknecht-Straße 24-25, 14476 Potsdam, Germany

Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Wilhelm-Conrad-Röntgen Campus, BESSY II, Albert-Einstein-Straße 15, 12489 Berlin, Germany

www.udkm.physik.uni-potsdam.de

*daniel.schick@uni-potsdam.de

Abstract: We present time-resolved X-ray diffraction data on antiferromagnetic Holmium thin films after direct femtosecond laser excitation. The strong magnetostriction in Holmium allows to correlate the observed ultrafast lattice dynamics to the antiferromagnetic helical spin structure.

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Metallic Holmium is a prototypical Heisenberg-antiferromagnet with a very large magnetic moment of $10.6\,\mu_B$ per atom, which is mainly due to the partially filled $4f$ shells. The indirect exchange interaction (RKKY) [1–3] leads to an incommensurate helical spin structure below the Néel temperature of approx. 131 K. [4] The strong magnetostriction in Holmium results in an increase of the c-axis [5] (negative thermal expansion coefficient) down to the ferro-to-antiferromagnetic phase transition at $T_C = 20\,\mathrm{K}$, cf. Fig. 1. The temperature-dependence of the spatial period of the spin helix in the antiferromagnetic phase of Holmium emphasizes the strong coupling between lattice and magnetic ordering. [6]

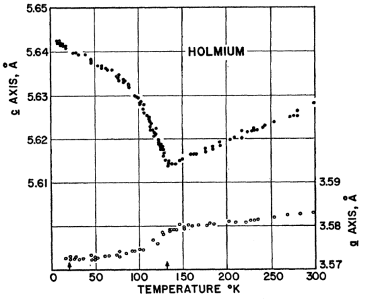

Fig. 1. Temperature-dependence of the a- and c-axis of Holmium reproduced from Ref. 5

We study a 80 nm $(0001)_{\mathrm{hcp}}$ Holmium film epitaxially grown on 70 nm $(0001)_{\mathrm{hcp}}$ Yttrium, a 50 nm $(110)_{\mathrm{bcc}}$ Niobium buffer layer, and a $(11-20)_{\mathrm{hcp}}$ sapphire substrate. The sample is cooled down to $T = 40\,\mathrm{K}$ well below the Néel temperature of Holmium. The ultrafast X-ray diffraction (UXRD) data are acquired by a laser-driven plasma X-ray source (PXS) [7] employing the ultrafast reciprocal space mapping technique. [8, 9] The static reciprocal space map (RSM) of the sample structure is shown in Fig. 2 on the left and features a significant mosaic broadening of the Yttrium, Holmium, and Niobium Bragg peaks.

The photoexcitation by 800 nm femtosecond laser pulses with an incident fluence of $F = 3.1\,\mathrm{mJ/cm^2}$ results in an ultrafast contraction of the Holmium layer within a few picoseconds with a maximum strain close to -0.3 % as shown in

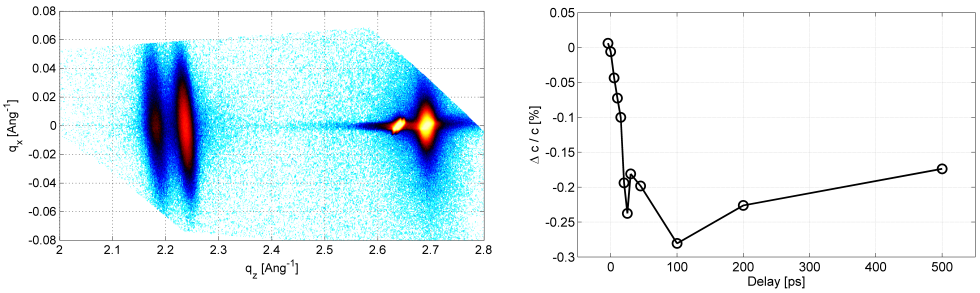

Fig. 2. (left) Static reciprocal space map (RSM) of the Holmium sample structure. From left to right the (0002) Bragg peaks of Yttrium and Holmium, as well as the (11-20) Sapphire substrate and (220) Niobium Bragg peaks. The instrumental function of the PXS diffractometer is indicated by the doubled and inclined Bragg peak of the Sapphire substrate. The other three materials exhibit large broadening along q_x likely due to mosaicity induced by the epitaxial mismatch. (right) The change of the c-axis lattice constant of Holmium is plotted as a function of delay time after photoexcitation ($t = 0$). The fast contraction of the Holmium lattice after photoexcitation is due to an ultrafast release of magnetostriction.

Fig. 2 on the right. The detailed evaluation of the in- and out-of-plane lattice dynamics provides important information about the spatio-temporal profile of the driving forces [9–11], such as the ultrafast modification of the spin structure in Holmium. The comparison of UXRD data with hard X-rays, which exclusively probe the atomic positions in the Holmium crystal, and time-resolved resonant magnetic soft X-ray scattering, which probes the correlation length and spatial period of the antiferromagnetic spin helix, allow to further disentangle the interplay between lattice degrees of freedom and magnetic exchange interaction on an ultrafast time scale.

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