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Resonant Magnetic Scattering in Holmium at an Undulator Source

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Abstract:

The resonance properties of the magnetic cross section of antiferromagnetic holmium were studied at the L absorption edges. A polarization analysis of the magnetic cross section was performed at the L_{III} and L_{II} edges using π polarized incident x-rays.

Magnetic x-ray scattering will benefit largely from the characteristics of the new high-brilliance synchrotron radiation sources (ESRF, APS and Spring8). Energy tunable undulators at ESRF provide a spectral brilliance of more than 10^{17} ph/s/0.1%bw/mm²/mrad² between 3 and 20 keV, a high degree of linear polarization ($P > 97\%$) and a low divergence (typically 20 μ rad) in both the vertical and horizontal planes. This allows quantitative polarization analysis of the magnetic scattering cross-section to be performed in both the resonant and non-resonant regimes and will lead to the determination of orbital- and spin magnetization densities in x-ray scattering experiments either via the polarization dependence of the non-resonant cross-section [1] or through the application of sum rules [2] to the dipolar contributions of the resonant magnetic cross-section obtained at two absorption edges. In addition it offers new possibilities in the basic determination of magnetic structures and in resonant spectroscopy.

We have undertaken the first magnetic scattering experiments at one of the new ESRF undulator beamlines (beamline 9 / Troika) and explored for the first time the polarization properties of the magnetic cross-section in antiferromagnetic holmium at the L_{III} and at the L_{II} absorption edge using a π polarized incident beam. Holmium is a spiral antiferromagnet ($T_N = 138K$) in which the average moments are ferromagnetically aligned within the basal planes, but rotate from plane to plane with an average turn angle proportional to the magnetic modulation wave-vector τ . When the energy of the incident x-rays is tuned

close to the L absorption edges a strong resonant enhancement of the magnetic scattering intensity is observed [3] and described using the X-Ray Resonance Exchange Scattering (XRES) model [4]. In holmium the resonant enhancements result from electric dipole- and quadrupole-like transitions with the sensitivity to the magnetization of the f- and d-bands mediated via the exclusion principle and spin-orbit correlations.

A holmium single crystal (mosaic width 0.03° at the (002) reflection) was supported in a displacive cryostat and studied with the (00L) face mounted in a horizontal reflection geometry. We studied the (002)+ τ magnetic satellite ($\tau=0.208c^*$ at 46.6 K) around the $L_I=9394$ eV, $L_{II}=8918$ eV and $L_{III}=8071$ eV absorption edges and observed strong resonant enhancements with counting rates of $>3 \times 10^5$ cps, 5×10^3 cps and 4×10^2 cps at the L_{III} , L_{II} and L_I edges respectively. Polarization analysis of the scattered intensity was carried out at the L_{III} and L_{II} absorption edges by means of PG(006) and Al(004) analyser crystals, respectively. The results (not corrected for absorption) are shown in figure 1. Figure 1 (top) shows the energy dependence of the non-rotated (π - π) and rotated (π - σ) component of the magnetic scattering intensity of the (002)+ τ satellite when the energy of the incident x-rays is tuned through the L_{III} absorption edge. The rotated component peaks above the L_{III} absorption edge and the lineshape of this resonance shows an asymmetric tail extending towards lower energy. The unrotated component (π - π) peaks below the edge and shows a slight asymmetry towards higher energies. Similar features were observed in holmium for incident σ polarization [3]. Although it is not possible to uniquely identify the multipole character (E1 or E2) of the two resonant components based on their polarization response (both polarization channels are allowed for both E1 and E2 transitions for incident π polarization) it seems reasonable in analogy to ref. [3] to interpret the rotated component (π - σ) peaking above the edge as arising from electric dipole transitions between the $2p_{3/2}$ state to the $5d$ band. The unrotated component results from quadrupolar transitions ($2p_{3/2} \leftrightarrow 4f$). The asymmetries can be interpreted as an interference between the resonant and non-resonant cross-sections. The most striking difference with the published data [3] is the magnitude of the unrotated (π - π) component relative to the π - σ component. This is consistent with the predictions for (π - π) scattering vs. (σ - σ) scattering given by the polarization term for the E2 scattering amplitude $i(k'k)(e'e)xz_j + i(e'e)(k'xk)z_j$ [4] with k, k' and e, e' being the (incident and scattered) wave- and polarization vectors, respectively.

The energy dependence of the magnetic scattering intensity at the L_{II} absorption edge is shown in figure 1 (bottom). We again observe resonant enhanced scattering in both polarization channels. The rotated component peaks above the absorption edge but there is a

very pronounced shoulder below the edge. The unrotated component is weaker and the data suggest a splitting with a component peaking above and one peaking below the edge. In analogy to the observations at the L_{III} edge we might tentatively interpret both components peaking above the edge as arising from E1 transitions and the components peaking below the edge as arising from E2 transitions, although both polarization channels are again allowed for both E1 and E2 transitions for incident π polarization.

Because the present data allow only a qualitative interpretation it is clear that an extended study at both absorption edges, including polarization analysis at the higher resonant harmonics, is needed in order to unambiguously identify the dipolar contributions for which the sum rule has been derived before being able to extract orbital and spin contributions to the magnetic moment.

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FIGURE CAPTIONS

Figure 1: Polarization dependence of the magnetic scattering intensity for the holmium (002)+ τ satellite at 46.6K as a function of the x-ray energy around the Ho L_{III} absorption edge (top) and the Ho L_{II} edge (bottom). The circles correspond to scattered σ polarization, the squares to scattered π polarization for a π polarized incident beam.

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