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Interpreting magnetic X-ray scattering on Gd-compounds using the McPhase simulation program

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Abstract

Magnetic X-ray scattering experiments on Gd-compounds provide valuable information about the magnetic structure, which is difficult to obtain by neutron scattering due to the large absorption cross-section of Gd. The interpretation of the results is usually difficult and sometimes ambiguous, and it would be desirable to have quantitative calculations in order to verify or disprove theoretical models by the data of scattering experiments. Such model calculations have been performed for the magnetic properties of the antiferromagnets $GdNi_2B_2C$ and $GdCu_2$. The results of these calculations are compared with experimental data from magnetic X-ray scattering experiments.

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1. Introduction

The magnetic anisotropy of rare-earth compounds usually is caused by the crystal field, unless the 4f angular momentum is zero ($L = 0$) such as in the case of Gd^{3+}. The small but finite magnetic anisotropy of $L = 0$ rare-earth compounds is reflected in the orientation of the moments with respect to the crystal lattice, which can be determined by magnetic scattering experiments. Because neutron diffraction on Gd compounds is difficult due to the large absorption cross-section of Gd, magnetic X-ray diffraction (MXD) using synchrotron light sources is a welcome alternative. The moment direction can be investigated using polarization analysis. The confrontation of these data with a quantitative model may lead to new insights about the magnetic interactions. Here we assume a simple model based on the following Hamiltonian:

$$\mathcal{H} = -\frac{1}{2}\sum_{ij}^{i\neq j}\left\{\mathcal{J}(ij)\mathbf{J}_i\mathbf{J}_j + \sum_{\alpha\beta} J_i^{\alpha}\mathcal{J}_{\alpha\beta}^{\mathrm{CD}}(ij)J_j^{\beta}\right\}, \quad (1)$$

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where the anisotropy enters only via the second term which corresponds to the classical dipole interaction

$$\frac{\mathscr{J}^{\mathrm{CD}}_{\alpha\beta}(ij)}{(g_J\mu_{\mathrm{B}})^2} = \frac{3(R^{\alpha}_i - R^{\alpha}_j)(R^{\beta}_i - R^{\beta}_j) - \delta_{\alpha\beta}|\mathbf{R}_i - \mathbf{R}_j|^2}{|\mathbf{R}_i - \mathbf{R}_j|^5}.$$

In these equations $\mathbf{R}_i$ denotes the lattice vector of the ith Gd ion, J^{α}_i ($\alpha = 1,2,3$) the three components of the total angular momentum, g_J the Landé factor, and μ_{B} the Bohr magneton. In the following we report the interpretation of MXD on two noncollinear antiferromagnets using a model analysis based on the Hamiltonian (1).

2. $GdCu_2$

$GdCu_2$ crystallizes in the orthorhombic $CeCu_2$ type of structure (space group Imma). At the Néel temperature of $T_{\mathrm{N}} = 41$ K the specific heat indicates an equal moment magnetic structure [1]. A noncollinear magnetic structure [2] was found by neutron diffraction. MXD [3] and μSR [4] agree with these results. The magnetostriction was interpreted quantitatively using correlation functions derived from the magnetic structure [5].

We have used MXD with polarization analysis to investigate the temperature dependence of the magnetic structure. In order to interpret our data a model calculation based on the Hamiltonian (1) was performed using the *McPhase* program [6, www.mcphase.de]. From the diagonal, but anisotropic exchange parameters found for $NdCu_2$ [7] only the isotropic part was taken and de Gennes scaled in order to obtain the exchange interaction for $GdCu_2$

$$\frac{\mathscr{J}^{\mathrm{GdCu_2}}(ij)}{(g^{\mathrm{Gd}}_J - 1)^2} = \frac{\sum_{\alpha=a,b,c}\mathscr{J}^{\mathrm{NdCu_2}}_{\alpha\alpha}(ij)}{3(g^{\mathrm{Nd}}_J - 1)^2}. \qquad (2)$$

In addition, the classical dipolar exchange for 1584 neighbors up to 3 nm was taken into account in the *McPhase* calculation.

At low temperatures, the calculated magnetic structure differs by a small tilt of the magnetic moments (8° in the AC plane) from the structure suggested by the neutron scattering experiments

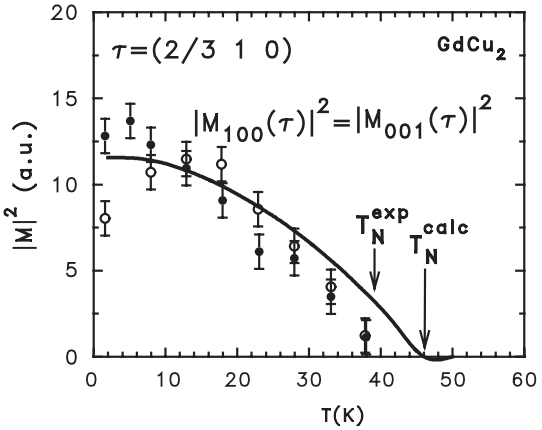

Fig. 1. $GdCu_2$—temperature-dependent measured MXD intensities on (3.6667 −1 1). Open (full) symbols correspond to $\pi\pi$ ($\pi\sigma$) MXD intensities, respectively. Lines correspond to the calculated temperature dependence of the squared FT of the magnetic moment configuration.

[2]. Such a tilt cannot be deduced with the neutron diffraction but was already suggested by μSR [4]. The temperature dependence of the Fourier transform (FT) of the magnetic moments $\mathbf{M}(\tau)$ has been computed and is compared in Fig. 1 to the temperature dependence of the MXD intensity measured on the main propagation vector with *ab* in the scattering plane. The temperature dependence of the $\pi\sigma$ intensity (sensitive to both M_{100} and M_{010}) agrees well with the calculated moment component along a (M_{100}), because the component M_{010} is zero at all temperatures (both in the calculation and as determined from the MXD results in the *ac* scattering plane [8]). The calculation predicts $M_{100} = M_{001}$ at all temperatures. Therefore it is difficult to understand the $\pi\pi$ data, which indicate, that M_{001} decreases below 10 K. Additional sources of anisotropy, other than the dipolar interaction have to be considered so as to account for this observation, which is also supported by a sharp increase of the susceptibility in c-direction below 10 K [3].

3. $GdNi_2B_2C$

$GdNi_2B_2C$ crystallizes in a body centered tetragonal structure (space group 14/mmm). The

Néel temperature is $T_N = 19.5$ K and a spin reorientation has been reported at $T_R = 13.5$ K. The magnetic structure has been investigated by MXD [9]. Above T_R the moments point along (010) with a propagation vector of $\tau = (0.55\,0\,0)$ while below T_R a (0 0 1) component of the moment appears. The propagation vector is only weakly temperature dependent (<0.5%).

For the *McPhase* calculation it is necessary to include 5 neighbors in order to get a global maximum of the FT of the isotropic interaction constants $\mathcal{J}(ij)$ at $\tau = (0.55\,0\,0)$. We used for the neighbors at $(0\,1\,0) - 12$ μeV, $(1\,1\,0) - 27$ μeV, $(0.5\,0.5\,0.5) - 29$ μeV, $(2\,2\,0) + 17$ μeV and at $(0\,0\,1) + 29$ μeV.

The experimentally observed magnetic structures including the moment directions are well reproduced by the calculation. The low-temperature modification of the magnetic structure deserves some comments. By MXD a spiral and a transverse wave were suggested as possible magnetic structures [9]. The *McPhase* calculation clearly stabilizes a spiral structure. Fig. 2 compares MXD data with the calculated temperature dependence of the FT of the magnetic moment **M**. Note that the measured resonant intensity corresponds to the component M_{001} (scaling with $|M_{001}|^2$), which appears below the spin reorientation temperature in accordance with the experiment [9]. The measured non-resonant intensity has to be compared to a linear combination of the M_{001} and M_{010} (see Fig. 2). The calculated longitudinal component M_{100} is zero at all temperatures. The factor of 1.986 in the linear combination is derived from $\cos^2(\Theta)/\sin^2(2\Theta)$. The calculation underestimates the stability range of the high temperature collinear magnetic structure. This temperature range is determined by the magnitude of the dipolar anisotropy, which confines the moments to the (0 1 0) direction.

In conclusion, a numerical calculation based on isotropic exchange and the dipolar interaction is able to predict the magnetic moment orientation for both systems under investigation. The remaining quantitative inconsistencies with the MXD experiment are attributed to anisotropic interactions, which are not taken into account in the model (1).

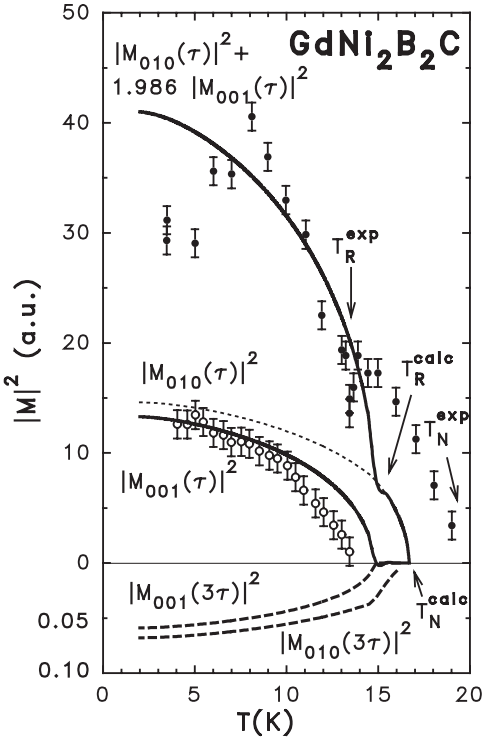

Fig. 2. Measured temperature dependence of the resonant (open symbols) and the non-resonant (full symbols) intensity of the first harmonic (1.447 0 0) magnetic reflection (data taken from [9] and scaled for comparison). Lines correspond to the calculated temperature dependence of the squared FT of the moment components M_{010} and M_{001} for the ordering wave vector τ and the harmonic 3τ.

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