Magnetic order in $GdMnO_3$ in high magnetic fields

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Abstract. Resonant magnetic x-ray scattering at the Gd L_2 edge is used to investigate the magnetic order of the Gd moments in multiferroic $GdMnO_3$ at low temperatures. We present high magnetic field data on the magnetic ordering of Gd in the ferroelectric phase of $GdMnO_3$. Our findings reaffirm the important role of the Gd moments in the symmetric magnetic exchange striction responsible for ferroelectricity in this compound.

1. Introduction

In recent years, strong magneto-electric coupling has been observed in orthorhombic rare-earth manganites $R\mathrm{MnO_3}$ [1, 2, 3, 4]. Ferroelectricity in these compounds is dependent on frustration-induced cycloidal ordering of the Mn magnetic moments, which breaks spatial inversion symmetry. This is however not the only prerequisite for multiferroicity, and possible microscopic mechanisms are still being explored [5]. The rare-earth moments have been observed to play a complex role in these materials. For example, in $DyMnO_3$ a threefold enhancement of the electric polarization is seen above 6.5 K, or when applying a magnetic field parallel to the a axis below 6.5 K, where Dy exhibits Mn-induced ordering [6, 7]. The basic mechanism for ferroelectricity in $DyMnO_3$ is *antisymmetric* exchange striction between neighboring Dy and Mn moments, which displaces the rare earth ion perpendicular to the ordering wave vector [8, 9, 10]. In contrast, the *symmetric* exchange striction is hypothesized to be responsible for the polarization enhancement in $DyMnO_3$, and also for the magnetic-field induced ferroelectricity in $GdMnO_3$ [11].

In $GdMnO_3$, both the Gd moments and the Mn moments order with propagation vector $\tau^{Gd} = \tau^{Mn} = \frac{1}{4}\mathbf{b}$ [7]. This can in theory cause a lattice modulation with period $q = \tau^{Gd} - \tau^{Mn} = 0$, i.e. a homogeneous lattice contraction or expansion capable of leading to ferroelectricity. However, in zero field $GdMnO_3$ is only ferroelectric in a small temperature range around 5 K, even though both Mn and Gd are ordered with $\tau = 1/4$ down to base temperature. Ferroelectricity is induced in a larger temperature range by application of a magnetic field along the b axis. Here, one paraelectric-to-ferroelectric transition coincides with a phase boundary within the Gd ordered state [7]. We have studied the Gd ordering as a function of temperature and magnetic field in order to shed some more light on the interplay between rare earth magnetism and ferroelectricity in $R\mathrm{MnO_3}$.

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2. Experiment

The experiment has been conducted at beamlines P09 at the PETRA III storage ring at Deutsches Elektronen-Synchrotron (DESY) [12] and 6-ID-B at the Advanced Photon-Source (APS), Argonne National Laboratory. The sample was mounted and cooled in a cryomagnet with field direction along the crystallographic b direction, perpendicular to the scattering plane. All measurements presented in this report were taken in the π-σ'polarization channel, unless otherwise noted.

The magnetic (0 1/4 6) reflection was investigated as function of temperature and magnetic field. At the Gd L_2 edge a strong resonant signal was measured in the π-σ'channel, confirming the magnetic origin of the reflection. At $T \sim 3K$ (not shown here), there is no intensity in the π-π'channel at resonance, consistent with Gd moments aligning along c as previously reported [7].

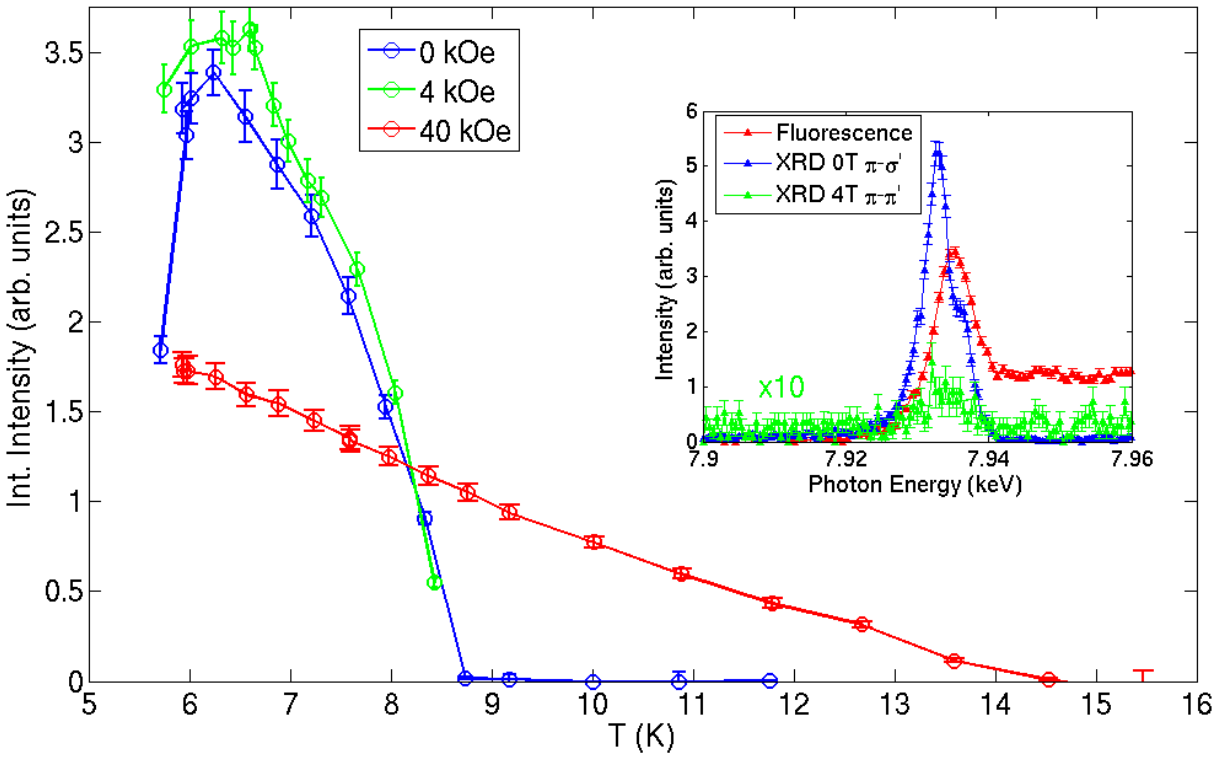

Figure 1. Temperature dependence of the (0 1/4 6) integrated intensity at different field values. The inset shows the energy dependence across the Gd L_2 edge at T = 5.7 K, in zero field in π-σ'as well as high field in π-π', along with a fluorescence measurement of the sample.

Figure 1 shows the dependence upon temperature of the magnetic scattering intensity, for different field strengths. In zero field, an intensity enhancement around $T \sim 6$K is seen. This is related to an increase of the b component of the Gd spins [7] and corresponds to the ferroelectric region in the low-field sector of the H-T phase-diagram from [4], ranging from about 5.5 K to 8.5 K. Imposing a weak field of 4 kOe results in a slight enhancement of the intensity in this region, while leaving the overall dependence intact. Increasing the field strength supresses the intensity again, but in turn extends the ordered phase to higher temperatures. With the field strength set to 40 kOe there is a weak XRD signal (which is clearly above the intensity level expected from cross-talk bwetween the polarization channels) also in the π-π'channel, indicating a small Gd spin component also in the b direction.

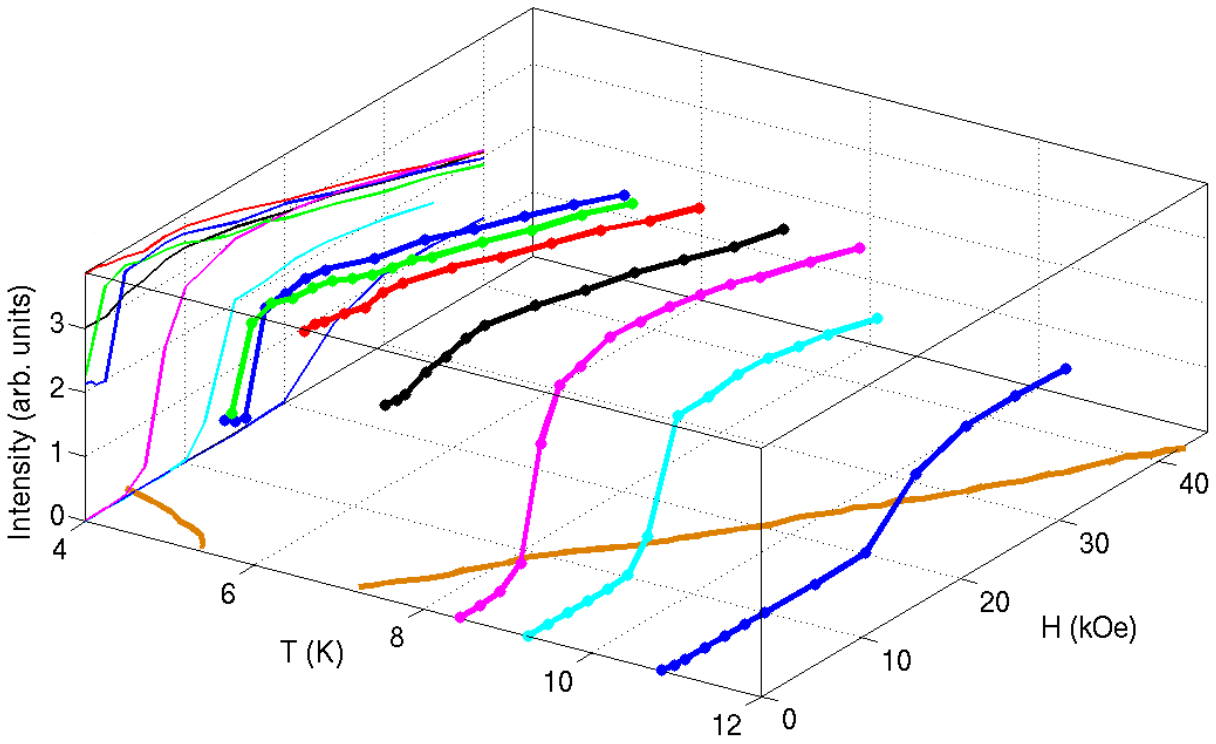

Figure 2. Field dependences of the (0 ¼ 6) integrated intensity as function of **H** ∥ **b** at fixed temperatures, with projections. The orange curves represent the boundaries of the ferroelectric phase from the phase diagram in [4].

In order to understand the effect of an imposed magnetic field, we recorded field dependences at several different values of constant temperature. The results are shown in figure 2. The measurements map out the H-T phase diagram of the Gd order. Comparing with the phase diagram from [4], one sees that the Gd ordered phase corresponds to the ferroelectric phase in all regions.

3. Discussion

The existence of a *b* component of the Gd moments in the high-field region is suggestive of an important role for the symmetric magnetic exchange interaction in stabilizing the ferroelectric phase. The symmetric exchange interaction between Gd and Mn moments is proportional to $\mathbf{S}_{Gd,i} \cdot \mathbf{S}_{Mn,j}$ which is zero when the Gd moments are oriented along *c*, perpendicular to the Mn moments in the *ab*-plane. Introducing a *b* component makes the exchange interaction non-zero, capable of causing a shift of the Gd ions towards the Mn ions. Our results therefore point to an important role of the rare earth moments for the ferroelectricity in orthomanganites, and may open up a simple route to enhance the ferroelectricity in multiferroics where the rare earth and transition metal ions order with the same wave vector.

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