Magnetic order in $GdMnO_3$ in magnetic fields

2014 J. Phys.: Conf. Ser. 519 012007

(http://iopscience.iop.org/1742-6596/519/1/012007)

Journal of Physics: Conference Series **519** (2014) 012007 doi:10.1088/1742-6596/519/1/012007

Magnetic order in $GdMnO_3$ in magnetic fields

A. Skaugen[1], D.K. Shukla[1,4], R. Feyerherm[2], E. Dudzik[2], Z. Islam[3], J. Strempfer[1]

[1] Deutsches Elektronen-Synchrotron DESY, 22603 Hamburg, Germany
[2] Helmholtz-Zentrum Berlin, BESSY II, 12489 Berlin, Germany
[3] Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439, USA
[4] UGC DAE Consortium for Scientific Research, Khandwa Road, Indore 01, India

E-mail: arvid.skaugen@desy.de

Abstract. Resonant magnetic x-ray scattering at the Gd L_2 edge is used to investigate the magnetic order of the Gd moments in the ferroelectric phase of multiferroic $GdMnO_3$ at low temperatures and under magnetic fields. 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 necessarily sufficient for creating multiferroicity, and additional 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]. On the other hand, ferroelectricity in $GdMnO_3$ is almost entirely dependent on an applied magnetic field. 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 $GdMnO_3$, the Mn moments order incommensurately below $T_N^{Mn} = 43$ K [2, 12]. Below the lock-in temperature, $T' = 23$ K, the Mn order undergoes a transition to commensurate A-type AF ordering associated with weak ferromagnetism [7]. This ordering breaks spatial inversion symmetry, which in similar orthomanganites leads to ferroelectricity at the gamma point[13]. In spite of this, $GdMnO_3$ is not ferroelectric in this phase, and it is necessary to look for additional mechanisms.

Below $T_N^{Gd} = 8.2$ K both the Gd moments and the Mn moments order with propagation vector $\boldsymbol{\tau}^{Gd} = \boldsymbol{\tau}^{Mn} = \frac{1}{4}\mathbf{b}^*$. This can in theory, through the symmetric exchange interaction, cause a lattice modulation with period $q = \tau^{Gd} - \tau^{Mn} = 0$, i.e. a homogeneous lattice contraction or expansion that under certain conditions may be polar, thus leading to ferroelectricity [7].

An overview of possible modulations arising from various types of spiral magnetic ordering has been listed by Arima *et al.* [14, 15]. However, in zero field $GdMnO_3$ is only ferroelectric in a small temperature range around 5 K, even though both Mn and Gd moments 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 magnetism at the Gd site and ferroelectricity in $GdMnO_3$.

2. Experiment

The experiment has been conducted at beamlines P09 at the PETRA III storage ring at Deutsches Elektronen-Synchrotron (DESY) [16] and 6-ID-B at the Advanced Photon-Source (APS) at Argonne National Laboratory. The sample was mounted and cooled in a cryomagnet with vertical field direction along the crystallographic b direction. The scattering plane was tilted slightly away from the ac plane in order to access reflections with a small k value. Unless otherwise noted, all measurements presented in this report were taken in the π-σ' polarization channel, where π (π') and σ (σ') denotes photon polarization of the incident (reflected) beam parallel and perpendicular to the scattering plane, respectively [17].

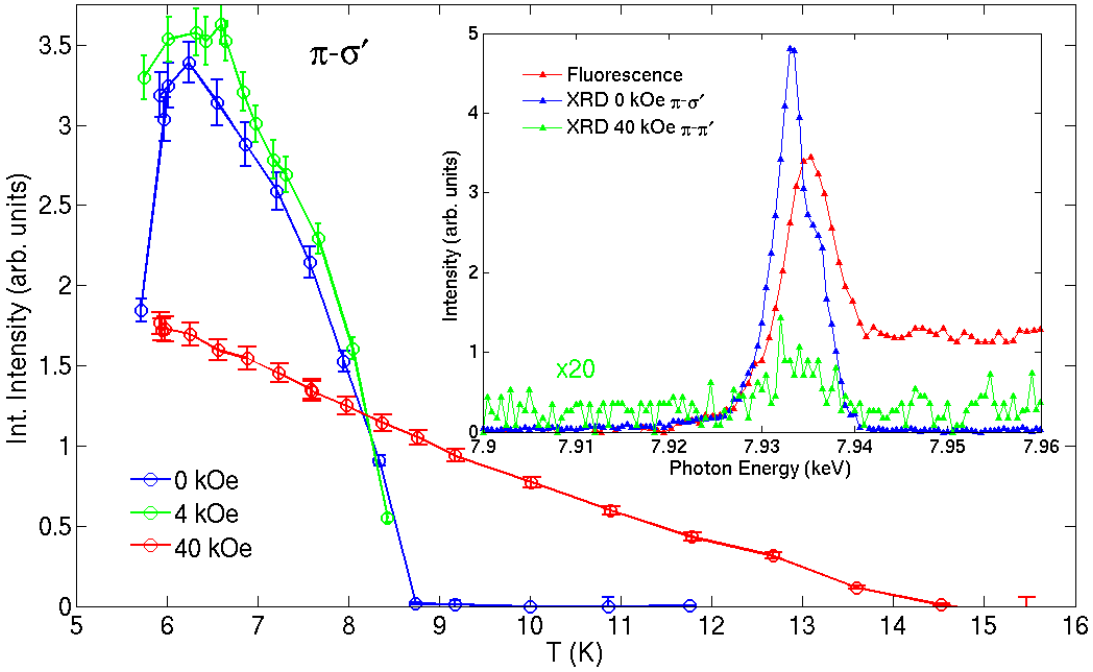

Figure 1. Temperature dependence of the (0 1/4 6) integrated intensity at different field values, measured in the π-σ' channel with a photon energy of 7.9325 keV. The inset shows the absorption corrected energy dependence across the Gd L_2 edge at T = 5.7 K, in zero field in π-σ' as well as under magnetic field in π-π', along with a fluorescence measurement of the sample.

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. Measurements were also performed with a perpendicular to the scattering plane. In this scattering geometry, at $T \sim 5.7$ K (not shown here), there is

no intensity in the π-π' channel at resonance, excluding any a component of the Gd moments. This is in accordance with previous studies indicating Gd ordering predominantly along c [7].

Figure 1 shows the dependence upon temperature of the magnetic scattering intensity in the π-σ' channel with b again perpendicular to the scattering plane, for different field strengths. In zero field, an intensity enhancement around $T^* \sim 6$ K is seen. Previous measurements have shown that 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. Application of a small field of 4 kOe results in a slight enhancement of the intensity in this region, while leaving the overall field dependence intact. Increasing the field strength to 40 kOe supresses the intensity again, but in turn extends the ordered phase to 15 K. This new transition temperature changes with field strength and does not correspond to any transition in the Mn order, indicating an onset of Gd self-ordering. 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 between the polarization channels) also in the π-π' channel, indicating a small Gd spin component also in the b direction.

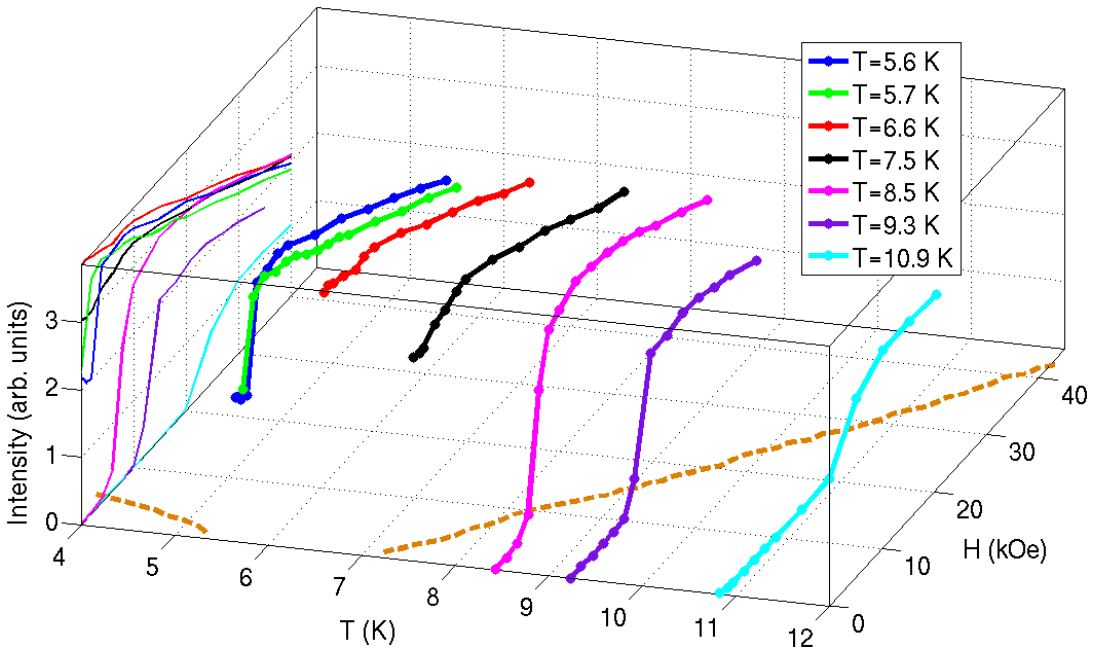

Figure 2. Field dependences of the (0 ¼ 6) integrated intensity as function of **H** ∥ **b** at fixed temperatures, with projections. The orange dashed curves represent the boundaries of the ferroelectric phase from the phase diagram in [4].

In order to understand the effect of an applied 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 Ref. [4], 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. Appearance of the b component at the Gd site 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.

Use of the Advanced Photon Source, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science by Argonne National Laboratory, was supported by the U.S. DOE under Contract No. DE-AC02-06CH11357.

References

[1] Fiebig M 2005 *J. Phys. D* **38** R123
[2] Kimura T, Goto T, Shintani H, Ishizaka K, Arima T and Tokura Y 2003 *Nature* **426** 55
[3] Goto T, Kimura T, Lawes G, Ramirez A P and Tokura Y 2004 *Phys. Rev. Lett.* **92** 257201
[4] Kimura T, Lawes G, Goto T, Tokura Y and Ramirez A P 2005 *Phys. Rev. B* **71** 224425
[5] Wang K F, Liu J M and Ren Z F 2009 *Advances in Physics* **58** 321–448
[6] Prokhnenko O, Feyerherm R, Dudzik E, Landsgesell S, Aliouane N, Chapon L C and Argyriou D N 2007 *Phys. Rev. Lett.* **98**(5) 057206 URL `http://link.aps.org/doi/10.1103/PhysRevLett.98.057206`
[7] Feyerherm R, Dudzik E, Wolter A U B, Valencia S, Prokhnenko O, Maljuk A, Landsgesell S, Aliouane N, Bouchenoire L, Brown S and Argyriou D N 2009 *Phys. Rev. B* **79**(13) 134426 URL `http://link.aps.org/doi/10.1103/PhysRevB.79.134426`
[8] Katsura H, Nagaosa N and Balatsky A V 2005 *Phys. Rev. Lett.* **95** 057205
[9] Sergienko I A and Dagotto E 2006 *Phys. Rev. B* **73**(9) 094434 URL `http://link.aps.org/doi/10.1103/PhysRevB.73.094434`
[10] Mostovoy M 2006 *Phys. Rev. Lett.* **96** 067601
[11] Feyerherm R, Dudzik E, Prokhnenko O and Argyriou D N 2010 *Journal of Physics: Conference Series* **200**(1) 012032
[12] Arima T, Goto T, Yamasaki Y, Miyasaka S, Ishii K, Tsubota M, Inami T, Murakami Y and Tokura Y 2005 *Phys. Rev. B* **72**(10) 100102 URL `http://link.aps.org/doi/10.1103/PhysRevB.72.100102`
[13] Aliouane N, Prokhnenko O, Feyerherm R, Mostovoy M, Strempfer J, Habicht K, Rule K C, Dudzik E, Wolter A U B, Maljuk A and Argyriou D N 2008 *J. Phys. Cond. Mat.* **20** 434215
[14] Arima T, Yamasaki Y, Goto T, Iguchi S, Ohgushi K, Miyasaka S and Tokura Y 2007 *J. Phys. Soc. Jpn.* **76** 023602
[15] Arima T 2007 *J. Phys. Soc. Jpn.* **76** 073702
[16] Strempfer J, Francoual S, Reuther D, Shukla D K, Skaugen A, Schulte-Schrepping H, Kracht T and Franz H 2013 *Journal of Synchrotron Radiation* **20** 541–549 URL `http://dx.doi.org/10.1107/S0909049513009011`
[17] Hill J P and McMorrow D F 1996 *Acta Crystallogr. A* **52** 236