Polarized-neutron-scattering studies on the chiral magnetism in multiferroic MnWO₄

T Finger¹, D Senff¹, K Schmalzl², W Schmidt², L P Regnault³, P Becker⁴, L Bohatý⁴ and M Braden¹

¹II. Physikalisches Institut, Universität zu Köln, Zülpicher Str. 77, D-50937 Köln, Germany
²Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, JCNS at ILL, 38042 Grenoble Cedex 9, France
³Institut Nanosciences et cryogénie, SPMS-MDN, CEA-Grenoble, DRFMC-SPSMS-MDN, F-38054 Grenoble Cedex 9, France
⁴Institut für Kristallographie, Universität zu Köln, Zülpicher Str. 49b, D-50674 Köln, Germany

E-mail: braden@ph2.uni-koeln.de

Abstract. Neutron diffraction with spherical polarization analysis is a powerful tool for studying the multiferroic materials where the ferroelectric polarization arises from a complex magnetic structure. Analyzing the off-diagonal terms in the polarization matrix one may directly detect the chiral contributions even in a multidomain arrangement. In MnWO₄ one can control the chiral magnetism by varying an electric field at constant temperature. The analysis of multiferroic hysteresis cycles at four equivalent magnetic Bragg peaks fully agrees with a nearly monodomain chiral arrangement controlled by the electric field. A pronounced asymmetry of the hysteresis cycles and memory effects point to strong pinning of the chiral magnetism in MnWO₄. We find a second-order harmonic modulation which exhibits both magnetic and structural character and which may be related with the domain pinning. The observed interference between the nuclear and the magnetic modulation is another manifestation of the coupling between the crystal structure and the magnetism in the multiferroic oxides.

1. Introduction

The complex magnetism in the recently discovered multiferroic transition-metal oxides attracts considerable attention, as these materials exhibit very large magnetoelectric effects possibly opening a route towards magneto-electric data-storage devices [1, 2, 3, 4, 5, 6]. In most of the newly discovered compounds, the multiferroic or magnetoelectric coupling arises from the asymmetric part of the magnetic interaction. In the same manner, as a low-symmetry crystal structure causes a canting of magnetic moments, the non-collinear magnetic order induces a structural distortion which may sum up into a ferroelectric polarization under the correct symmetry. Following references [7, 8, 9] the induced electric polarization of a single pair of spins $S_i, S_j$ separated by a distance vector $r_{i,j}$ is given by: $P_{FE} \propto r_{ij} \times (S_i \times S_j)$. For this inverse Dzyaloshinskii-Moriya (IDM) mechanism a non-collinear magnetic structure is a necessary condition rendering longitudinal and spherical polarization analysis a powerful and nearly unique method [10].

In the REMnO₃ series, depending on temperature and on the type of the RE, several magnetic-field induced effects are described: One may induce a finite ferroelectric polarization...
out of a paraelectric phase, one may suppress a ferroelectric polarization existing at zero magnetic field, or one may flop a ferroelectric polarization into another direction [3, 6, 11]. However, the opposite sense of the multiferroic coupling is more demanding to study, as the complex magnetic ordering requires a microscopic probe. A circular cycloid magnetic structure yields optimum conditions in view of the generation of ferroelectric polarization. A magnetic cycloid does not possess a scalar chirality or handedness (for example defined as $q_{\text{prop}} \cdot (S_1 \times S_2)$), since the propagation vector is parallel to the spiral plane of the magnetic moments. However, the IDM mechanism involves the cross product of neighboring spins and can, therefore, be associated with the chiral term:

$$M_{\text{ch}}(Q) = i \{M_{\perp}(Q) \times M_{\parallel}^*(Q)\},$$

entering the polarized-neutron-scattering cross sections [10].

By poling the multiferroic compounds in moderate magnetic fields it was possible to prove by polarized neutron scattering, that the sign of the chiral term follows the sign of the poling electric field in several transition-metal oxide multiferroics: in TbMnO$_3$ [12], in LiCu$_2$O$_2$ [13] and in MnWO$_4$ [14]. This magnetic poling gives strong support to the IDM multiferroic mechanism [7, 8, 9]. The IDM coupling is, however, a second-order effect, which, in addition to strong frustration as cause of the complex magnetism, yields only a limited amount of ferroelectric polarization. In the REMnO$_3$ [3, 6, 15] series the polarization is about two orders of magnitude smaller than in a typical perovskite ferroelectric [15]. Other materials, like MnWO$_4$ [16, 17, 18] or the pyroxenes [19] exhibit a ferroelectric polarization reduced by more than three orders of magnitude compared to BaTiO$_3$ [3, 15] hindering the observation of electric-field induced effects in the magnetic structure as well as applications.

In our recent work we have studied MnWO$_4$ where the multiferroic poling exhibits a memory effect which is not erased upon heating deeply into the paramagnetic and paraelectric phase [20]. In addition it has been possible to switch the chiral magnetism by varying the applied electric field at constant temperature, which is the relevant effect in view of potential applications. Here we extend our work by further hysteresis-curve measurements and by an analysis of the second-order harmonic modulation, which in MnWO$_4$ has both nuclear and magnetic character.

The crystal structure of MnWO$_4$, see Figure 1, is characterized by zigzag chains formed by the MnO$_6$ octahedra [21] (space group P2/c, $a=4.835\,\text{Å}$, $b=5.762\,\text{Å}$, $c=4.992\,\text{Å}$ and $\beta=91.08^\circ$) suggesting a one-dimensional magnetic character. However, the analysis of the spin-wave dispersion [22] finds strong interchain interactions indicating that the magnetic character in this material is not one-dimensional but frustrated. Upon cooling, MnWO$_4$ exhibits a first transition into an incommensurate magnetic phase labelled AF3 with propagation vector $q_{\text{ic}}=(-0.214,0.5,0.457)$ and collinear moments aligned in the $a,c$-plane, $T_{AF3}=13.2\,\text{K}$ [21, 22] followed by a second transition to a still incommensurate but non-collinear phase, AF2, this magnetic structure is displayed in Figure 1. In the AF3 phase the magnetic moment aligns along an easy axis, $e_{\text{easy}}$, in the a,c-plane which forms an angle to the propagation vector $q_{\text{ic}}$ of $83^\circ$. The transition into the AF2 phase is nearly continuous and can be characterized by the continuous growing of the b-component of the ordered moment, see Figure 1 and reference [20]. The diffraction studies [21] were unable to fix the phase between the easy-axis and the b components of the AF2 structure, but a chiral arrangement was indirectly deduced from the size of the moments. The transition AF3→AF2 is accompanied by the occurrence of the ferroelectric polarization [16, 17, 18]. Finally, the magnetic structure transforms into a commensurate collinear AF1 state with $q_c=(-0.25,0.5,0.5)$ at $T_{AF1}=7.0\,\text{K}$, where the ferroelectric polarization disappears and where magnetic moments again exhibit a collinear arrangement aligned along $e_{\text{easy}}$. The AF1 magnetic structure seems to be very anharmonic [21].
2. Polarized-neutron-scattering experiments on MnWO\(_4\)

2.1. Experimental

We have performed the polarized-neutron-diffraction studies on the IN12 cold triple-axis spectrometer at the Institut Laue-Langevin using the Cryopad-equipment for spherical polarization analysis [10]. A supermirror bender and a (111)-Heusler crystal were used for the polarization control on the \(k_i\) and \(k_f\) sides, respectively. We use the common cartesian coordinate system with \(x\) along \(Q\), \(y\) in the scattering plane but perpendicular to \(Q\), and \(z\) vertical. In our notation \(\sigma^{ij}_{\downarrow\uparrow}\) denotes the intensity in the channel with the outgoing polarization along \(j\) when the incoming polarization is set along \(i\) with the arrows indicating the directions of polarizations.

With the experimental setup we achieved flipping ratios of 35 and 40 in the two experimental runs, as measured on a nuclear Bragg peak. The precision of the spherical analysis was checked on the purely nuclear (020) Bragg reflection, where no rotation of polarization should occur. At the (020) reflection the rotation of the neutron polarization is very small underlining the precision of the neutron polarimetry. The polarization in the three longitudinal (diagonal) channels amounts to 94.5%; in the other channels we find: \(xy\) 4.9%, \(xz\) 1.2%, \(yx\) 3.8%, \(yz\) 0.7%, \(zx\) 1.6%, and \(zy\) 2.3%. These polarizations of the outgoing beam show that the neutron polarization can be controlled within a precision of about a degree, just in the \(xy, yx\)-channels a still small erroneous leakage seems to occur. In order to access the chiral terms in the incommensurate magnetic phases we had to use the unconventional \((0, 1, 0), (-0.214, 0, 0.457)\) scattering plane using a synthetic single crystal [16]. At the magnetic Bragg peaks \((-0.214, 0.5, \pm 0.457)\) and \((\pm 0.214, -0.5, \pm 0.457)\) we find a flipping ratio in the \(xx\) channel of 33 to be compared to the value of 35 found at the purely nuclear Bragg peak in the same run. This measurement shows that at the first-order magnetic Bragg peak there is only a negligible nuclear contribution of less than \(5 \cdot 10^{-3}\) and that the orientation of the polarimetry coordinate system versus the scattering vector is very precise. The electric field was always applied parallel to the \(b\)-direction by placing the plate-shaped crystal of 4mm thickness between Al-plates which are nearly transparent for neutrons.

Figure 1. (a) Crystal structure in MnWO\(_4\) with chains of MnO\(_6\) octahedra running along the \(c\) direction; b) the magnetic structure of MnWO\(_4\) in the incommensurate chiral phase AF2; the blue arrow indicates the magnetic propagation vector which is almost perpendicular to \(\mathbf{e}_{\text{easy}}\). c) temperature dependence of the chiral scattering in MnWO\(_4\) obtained in opposed electric poling fields at the first-order magnetic Bragg peak \((0.214, 0.5, -0.457)\). The field was applied along the \(b\)-direction. The chiral contribution only roughly scales with the electric polarization taken from reference [17] due to a minor amount of misaligned chiral domains, which furthermore can be temperature dependent.
2.2. Poling of chiral magnetism

In the used scattering plane, the easy-axis of MnWO$_4$ points almost perfectly along the $z$-direction. Therefore, the spin-flip scattering in the longitudinal (diagonal) channels along $y$ and $z$ sense the $e_{\text{easy}}$ and $b$ components of the magnetic order, respectively. Following the temperature dependencies of the $\sigma_{yy}^{\uparrow\downarrow}$ and the $\sigma_{xx}^{\uparrow\downarrow}$ spin-flip scattering one measures the appearance of the magnetic $b$-component in the AF2 phase and its suppression in the AF1 phase at low temperature [20], note that the propagation vector is almost perpendicular to $e_{\text{easy}}$. In addition, the chiral contribution can be effectively measured at $Q=(-0.214,0.5,0.457)$ and $(-0.214,1.5,0.457)$. The chiral term contributes to three channels in spherical polarization. In comparison to the total magnetic scattering it is measured in the $\sigma_{yy}^{\uparrow\downarrow}$ and $(-0.214,1.5,0.457)$. The chiral term contributes to three channels in spherical polarization.

The chiral contribution, $M_{\text{ch}}(Q)$, and the chiral ratio, $r_{\text{chir}}$, can also be measured at the non-diagonal components of the polarization tensor:

$$r_{\text{chir}} = \frac{M_{\text{ch}}(Q)}{M_y(Q) \cdot M_y^*(Q) + M_z(Q) \cdot M_z^*(Q)} = \frac{\sigma_{xx}^{\uparrow\downarrow} - \sigma_{xx}^{\uparrow\downarrow}}{\sigma_{xx}^{\uparrow\downarrow} + \sigma_{xx}^{\uparrow\downarrow}}. \quad (2)$$

In the $yx$ and similarly in the $zx$ channels [10]. Since the studied magnetic Bragg peaks (besides the second-order peaks, see below) are purely magnetic we may fully neglect any nuclear term and its interference with the magnetism. The polarization matrices measured at the purely magnetic Bragg peaks further document the high polarization precision, as the $xy$ and $xz$ channels exhibit polarizations of only 3%.

Similar to earlier observations on this and other chiral multiferroics it is possible to pole the chiral component with an electric field [12, 13, 14, 20]. In MnWO$_4$ we observe, after cooling in an electric field of 3500V/4mm, a chiral ratio of 81% at $Q=(-0.214,0.5,0.457)$ and $T=7.7$ K [20] which is slightly below the expected value. Supposing that the phase between the $e_{\text{easy}}$ and the $b$ components is 90° and that the spiral is perfectly circular, one calculates an expected chiral ratio of 96%. This value still deviates from one due to the projections of the magnetic moment to the $(x,y,z)$ coordinate system defined above. There are several reasons which may explain the reduced experimental chiral ratio. First the imperfect polarization reduces the measured chiral ratio. In addition, we find that the neutron beam becomes slightly depolarized for initial polarization along $y$ and $z$. From that and the polarization corrected chiral ratio we may conclude that about 5% of the sample volume remain in the wrong chiral arrangement [20]. Taking further into account that the two magnetic components are of equal size [20], we may further conclude that the chiral structure of MnWO$_4$ is close to ideal, i.e. circular and with a 90° phase shift between the components. The relative amount of the misaligned domains may vary as function of temperature in the used large sample crystal explaining the limited scaling between the chiral contribution and the ferroelectric polarization, shown in Figure 1. Indeed a better description of the ferroelectric polarization is obtained for plotting the product of the two magnetic components, see reference [20], indicating that the phase of the magnetic structure amounts to about 90° throughout the range of the AF2 phase.

2.3. Electric-field driven hysteresis curves

The control of magnetism by an external field at constant temperature is most important in view of the desired future applications, but only very little experiments of this type have been reported...
in general [23, 24, 25, 26, 27, 28]. In particular the electric-field control of the chiral magnetism in the multiferroics driven through the IDM mechanism has not been studied until recently. In MnWO₄ it has been possible to switch the electric polarization in thin crystals by varying the electric field [29]. From this effect one may indirectly deduce via the IDM mechanism a switching of the chiral domains. In addition, studies of the second-harmonic generation find a switching of magnetic domains induced by an electric field [30]. A direct observation of multiferroic hysteresis cycles was obtained by the polarized neutron diffraction experiments [20], where it was possible to control the chiral ratio defined in equation (2,3) by varying the electric field. After completion of this work we learned that a similar hysteresis curve was also reported in [31].

![Figure 2. Hysteresis curves obtained by measuring the chiral ratio as a function of external electric field at a constant temperature of T=10.5 K: nearly identical hysteresis cycles were obtained at the four equivalent Q-positions (±0.214,0.5,±0.457) and (±0.214,-0.5,±0.457) studied.](image)

All the hysteresis cycles reported in reference [20] were recorded at Q= (-0.214,+0.5,+0.457). Here we present in addition the cycles at the symmetrically equivalent Q-positions. Between these measurements the sample was always heated to 15 K and cooled with a voltage of -3500V. The hysteresis cycles shown in Figure 2 were then recorded by varying the voltage from -3500V to +3500V and back to -3500V. Due to the monoclinic symmetry one may only invert the signs of the qₖ and qᵢ components simultaneously in order to reach an equivalent position in reciprocal space, corresponding to an inversion of the incommensurate part of the propagation vector. The inversion of qₖ is irrelevant, as k=±0.5 generates the same magnetic structure with antiferromagnetic coupling of neighboring chains along the b direction (note that the difference k = ±0.5 is just a reciprocal lattice vector). The real crystal consists of domains corresponding to (±0.214,0.5,±0.457) modulation vectors which are associated with different signs of the chiral component at a given scattering vector, see equation (1). Alternatively, one may consider the chiral domains as arising from an inversion of one of the magnetic components, either that along b or that along e_easy. One of these chiral arrangements is shown in Figure 1. The real domain structure of this magnetic configuration may be more complex due to phase shifts of the magnetic moments. The four hysteresis-cycles recorded at T=10.5 K at the four Q-positions (±0.214,0.5,±0.457) and (±0.214,-0.5,±0.457) are almost perfectly identical thereby documenting the reproducibility of the setup. When analyzing the inverted Q-position in this nearly perfect chiral mono-domain structure, one might expect to find the scattering in the reverted channel, σ₁₁ versus σ₁₂. However, one has to keep in mind that the (x,y,z) coordinate system is fixed to the scattering vector, and that it is rotated by 180° in respect with the crystal coordinate system by the inversion of the scattering vector. For a given chiral domain, we measure thus the identical chiral ratio at the (±0.214,±0.5,±0.457) scattering vectors. The
simple inversion of \( q_k \) does not change neither the incommensurate part of the propagation vector nor the orientation of the \((x, y, z)\) coordinate system with respect to the magnetic components projected perpendicular to the scattering vector. Therefore, there is again no reversal of the measured chiral ratios, so that all four hysteresis curves are almost identical. Minor differences arise from the difficulty to fully reproduce the initial conditions and the different orientation of the large crystal in the neutron beam. We may conclude that the hysteresis cycles at the four equivalent magnetic Bragg positions perfectly agree with a nearly monodomain chiral structure, which can be inverted by the application of an external electric field.

2.4. Second-order anharmonic modulation

The poling of the chiral component exhibits a complex memory effect in MnWO\(_4\) [20]. The crystal remembers a preference for a certain chiral arrangement even when heating deeply into the paramagnetic and paraelectric phase. A related memory effect has also been observed when cycling the sample across the paraelectric AF1 phase [32] and when suppressing the ferroelectric AF2 phase by a magnetic field [30]. The latter study of the second-harmonic generation nicely documents that the identical domain structure is recovered when suppressing the AF2 phase by a magnetic field and setting the magnetic field back to zero. In particular our observation that the memory is not erased upon heating up to 30 K renders the given interpretation [32] in terms of multiferroic embryos very unlikely. We interpret the memory due to the pinning of the ferroelectric and magnetic domain walls by defects. Furthermore, relaxation processes similar to those reported for REMnO\(_4\) might be involved in the memory effects [33].

If the ferroelectric structure is purely electronic in origin, it appears difficult to explain a strong magneto-elastic pinning. Due to the weakness of the ferroelectric polarization, even a nuclear ferroelectric origin might be too weak in order to cause an efficient pinning. We have, therefore, searched for other processes pinning the multiferroic phase by analyzing the scattering

Figure 3. Analysis of the scattering at the second-order harmonic reflection (-0.428,-1,0.914); part a) and b) show scans across the reflection in transversal (\( \omega \)-scan) and [010] direction, respectively for spin-flip and non-spin-flip scattering in the \( xx \) channel; part c) presents the temperature dependencies of the magnetic and nuclear contributions across the magnetic phases of MnWO\(_4\).
at twice the propagation vector in the AF2 and AF3 phases. The results are given in Figure 3. We have studied the second-order modulation at $Q = (-0.428, 1.0, 0.914)$ finding a sizeable signal. Already in the collinear AF3 phase we find intensity in the non-spin-flip channels $\sigma_{xx}$. An incommensurate collinear magnetic structure is always coupled to a structural modulation of half the period in real space (and twice the propagation vector) due to the exchange striction modulating the magnetic interaction. In the AF2 phase this second-order harmonic scattering seems to increase much more than one might expect from the total increase in the ordered moment. Furthermore, the usual exchange-striction mechanism should not apply in an ideal chiral magnetic structure as the scalar product of coupled spins, $S_i \cdot S_j$, along the propagation vector does not vary along the propagation vector. Most interestingly we also find a magnetic component at the position of the second-order harmonics, which is limited to the AF2 phase. The magnetic second-order scattering is, however, three orders of magnitude smaller than the first-order magnetic scattering. Therefore, the anharmonic modulation of the magnetic structure in MnWO$_4$ is only a minor distortion. The ferroelectric chiral AF2 phase exhibits thus a $2 \cdot q_{ic}$ modulation of both the magnetic and the crystal structure. These anharmonic modulations might be key elements for the understanding of the anomalous memory and asymmetry effects in MnWO$_4$ and in other multiferroics.

The measurement of the full polarization matrix at $Q = (-0.428, 1.0, 0.914)$ gives further insight into the nature of the second-order modulation: There is a strong nuclear-magnetic interference term indicating that the magnetic and nuclear modulations are coexisting and intimately coupled. The real part of the nuclear-magnetic interference $R_i$ is determined in the longitudinal (diagonal) channels through [10]: $R_i = \frac{\sigma_{||}^{\uparrow\downarrow} - \sigma_{||}^{\downarrow\uparrow}}{2}$. We find $R_y = 1738 \pm 226$ and $R_z = 378 \pm 1263$ counts, far beyond statistical errors. Similarly significant evidence is also found in the non-diagonal channels $xz$, $yz$ and $xy$, $zy$ indicating a dominating real part of the nuclear magnetic interference. The nuclear-magnetic interference clearly documents that the second-order harmonic distortions are closely coupled. Thereby the chiral magnetism gets fixed to an incommensurate lattice modulation. Since the latter can be effectively pinned by defects or by an extrinsic distortions, the entire multiferroic domains will be pinned much more effectively than one what one would expect for just a ferroelectric structural modulation.

3. Conclusion

Neutron diffraction with spherical polarization analysis is a very powerful tool for studying the multiferroic transition-metal oxides, where ferroelectric coupling arises from a chiral magnetic structure. Analyzing the diagonal and off-diagonal terms one may directly access the chiral arrangement in a mono-domain sample, and the depolarization of the beam can point to strong chiral components even in a chiral multi-domain arrangement [10].

In MnWO$_4$ we find the close to perfect poling of the chiral components in large single-crystals, which exhibits an anomalous memory effect. Detailed analysis of the spherical polarization indicates that MnWO$_4$ at low temperature in the AF2 phase exhibits a nearly perfect circular chiral state. The product of the magnetic components scales very well with the ferroelectric polarization giving further support to the IDM mechanism.

The chiral arrangement can be controlled at constant temperature by varying the electric field [20], which corresponds to the effect desired for applications. However, the hysteresis cycles are broad and asymmetric due to an effective pinning of the domains in the chiral ferroelectric phase. Hysteresis cycles at four equivalent $Q$-positions agree perfectly to each other and to the expectations for a nearly monodomain chiral phase, which can be inverted by the application of an external electric field.

As a possible origin of the pinning of the multiferroic phase, we may identify a structural and magnetic $2 \cdot q_{ic}$ modulation. In the chiral ferroelectric AF2 phase, MnWO$_4$ exhibits an anharmonic magnetic modulation detected at the second-order scattering position. This
magnetic anharmonicity seems to disappear in the collinear AF3 phase. In the AF3 and in the AF2 phase there is in addition a structural modulation characterized by the doubled modulation vector, which however, is much stronger in the ferroelectric AF2 phase. The structural and the second-order harmonic magnetic modulations in the multiferroic AF2 phase seem to be relevant for the understanding of the pinning of magnetic and ferroelectric domains. The observation of strong nuclear-magnetic interference terms at the second-order peak is another manifestation of the strong coupling between nuclear structure and magnetism in the multiferroic materials.

Acknowledgments

This work was supported by the Deutsche Forschungsgemeinschaft in the Sonderforschungsbereich 608.

References

[1] Eerenstein W, Mathur N D and Scott J F 2006 *Nature* **442** 759
[2] Cheong S-W and Mostovoy M 2007 *Nature Materials* **6** 13
[3] Kimura T, Ishihara S, Shintani H, Arima T, Takahashi K, Ishizaka K and Tokura Y 2003 *Nature* **426** 55
[4] Hur N, Park S, Sharma P, Ahn J, Guha S and Cheong S-W 2004 *Nature* **429** 392
[5] Lawes G, Harris A B, Kimura T, Rogado N, Cava R J, Aharony A, Entin-Wohlman O, Yildirim T, Kenzelmann M, Broholm C and Ramirez A P 2005 *Phys. Rev. Lett.* **95** 087205
[6] Goto T, Kimura T, Lawes G, Ramirez A and Tokura Y 2004 *Phys. Rev. Lett.* **92** 257201
[7] Katsura H, Nagaosa N and Balatsky A 2005 *Nature* **426** 55
[8] Hur N, Park S, Sharma P, Ahn J, Guha S and Cheong S-W 2004 *Nature* **429** 392
[9] Mostovoy M 2006 *Phys. Rev. Lett.* **96** 067601
[10] Chatterji T 2006 *Neutron Scattering from Magnetic Materials*, Elsevier, Amsterdam
[11] Aliouane N, Schmalzl K, Senff D, Maljuk A, Prokes K, Braden M and Argyriou D N 2009 *Phys. Rev. Lett.* **102** 207205
[12] Yamazaki Y, Miyasaka S, Kaneko Y, He J-P, Arima T and Tokura Y 2007 *Phys. Rev. Lett.* **98** 147204
[13] Seki S, Yamazaki Y, Soda M, Matsuura M, Hirota K and Tokura Y 2008 *Phys. Rev. Lett.* **100** 127201
[14] Sagayama H, Taniguchi K, Abe N, Arima T, Soda M, Matsuura M and Hirota K 2008 *Phys. Rev. B* **77** 220407
[15] Kimura T, Lawes G, Goto T, Tokura Y, and Ramirez A 2005 *Phys. Rev. B* **71** 224425
[16] Heyer O, Hollmann N, Klassen I, Jodlauk S, Becker P, Mydosh J A, Lorenz T and Khomskii D 2006 *J. Phys. Condens. Matter* **18** L1471
[17] Taniguchi K, Abe N, Takenobu T, Iwasa Y and Arima T 2006 *Phys. Rev. Lett.* **97** 097203
[18] Arkenbout A H, Palstra T T M, Siegrist T and Kimura T 2006 *Phys. Rev. B* **74** 184431
[19] Jodlauk S, Becker P, Mydosh J A, Khomskii D I, Lorenz T, Streltsov S V, Hezel D C and Bohatý L 2007 *J. Phys. Condens. Mater* **19** 423201
[20] Finger T, Senff D, Schmalzl K, Schmidt W, Becker P, Bohatý L and Braden M arXiv:0907.5319
[21] Lautenschlager G, Weitzel H, Vogt T, Hock R, Bonnet M and Fuess H 1993 *Phys. Rev. B* **48** 6087
[22] Ehrenberg H, Weitzel H, Fuess H and Hennion B 1999 *J. Phys. Condens. Matter* **11** 2649
[23] Asher E, Rieder H, Schmid H and Stössel H 1966 *J. of Appl. Physics* **37** 1404
[24] Lottermoser T, Lonkai Th, Amann U, Hohlwein D, Ihringer J and Fiebig M 2004 *Nature* **430** 541
[25] Bodenthin Y, Staub U, García-Fernández M, Janoschek M, Schlappa J, Golovchets I E, Sanina V A and Lushnikov S G 2008 *Phys. Rev. Lett.* **100** 027201
[26] Zhao T, Scholl A, Zavaliche F, Lee K, Barry M, Cruz M P, Chu Y H, Ederer C, Spaldin N A, Das R R, Kim D M, Baek S H, Eom C B, Golovenchits E I, Sanina V A and Fiebig M 2009 *Nature Materials* **5** 823
[27] Radaelli P G, Chapon L C, Daoud-Aladine A, Vecchini C, Brown P J, Chatterji T, Park s and Cheong S W 2008 *Phys. Rev. Lett.* **101** 067205
[28] Saito M, Ishikawa K, Konno S, Taniguchi K and Arima T 2009 *Nature Materials* **8** 634
[29] Kundys N, Simon C and Martin C 2008 *Phys. Rev. B* **77** 172402
[30] Meier D, Maringer M, Lottermoser T, Becker P, Bohatý L and Fiebig M 2009 *Phys. Rev. Lett.* **102** 107202
[31] Poole A, Brown P J and Wills A S 2009 *J. Phys. Conf. Series* **145** 012074
[32] Taniguchi K, Abe N, Ohtani S and Arima T 2009 *Phys. Rev. Lett.* **102** 147201
[33] Schrettle F, Lunkenheimer P, Hemberger J, Ivanov V Yu, Mukhin A A, Balbashov A M and Loidl A 2009 *Phys. Rev. Lett.* **102** 207208