Field-induced chirality in the helix structure of Ho/Y multilayers

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We study the net chirality in the spin helix structure of Ho/Y multilayers induced by an in-plane applied magnetic field. The lifting of degeneracy of the chiral symmetry was revealed by means of polarized neutron reflectometry. Three samples of different thicknesses of the Ho and Y layers were grown by molecular-beam epitaxy. The chiral states are degenerated upon zero field cooling below the critical temperature $T_N = 115 \pm 3$ K. The chirality parameter γ rises during the field cooling procedure in the field range from 0 to 1 T and saturates at a value of 0.12 ± 0.01. The chirality appears stepwise below T_N and depends weakly on temperature. The phenomenon is interpreted in terms of the Dzyaloshinskii-Moriya interaction appearing at the interface between the Ho and Y layers.

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I. INTRODUCTION

Rare-earth magnetism has attracted much attention in light of the discovery of three-dimensional (3D) long-range order, which can occur in rare-earth/yttrium superlattice (SL) structures [1–6]. Superlattices of Dy/Y and Ho/Y show a helical order in which the magnetic moments are aligned in ferromagnetic sheets within each basal plane, but the orientation of these moments changes from one plane to another, thus forming a spin helix. The long-range coherence of the magnetic structure arises from the conduction electrons propagating coherently throughout the SL. This coherent propagation from the yttrium layers into the magnetic layers maintains the stability of the turn angle and the chirality of the helix.

The most recent impulse for investigations of the rare-earth superlattice was stimulated by the observation of effects caused by magnetic interfaces, such as enhanced interfacial magnetic order [7], twisted magnetization states [8], and the surface-induced Dzyaloshinskii-Moriya interaction [9–11]. A few years ago Grigoriev *et al.* demonstrated that Dy/Y magnetic multilayers (MMLs) possess a coherent spin helix with a preferable chirality induced by a magnetic field [10]. It was shown that a magnetic field applied in the plane of the sample upon cooling below T_N is able to repopulate the otherwise equal population numbers for the left- and right-handed helixes. It was suggested that the interplay of the Ruderman-Kittel-Kasuya-Yoshida (RKKY) and Zeeman interactions helps to reveal the otherwise hidden antisymmetric Dzyaloshinskii-Moriya interaction (DMI). It was argued that the observed chirality is a fingerprint of the DMI resulting from a lack of symmetry inversion at the interfaces [12].

One can suggest that the same effect of an applied magnetic field on the chirality of the helix spin structure can occur in the MMLs made of other rare-earth elements such as Ho. The magnetic structure of bulk Ho was investigated using neutron scattering by Koehler *et al.* [13]. Below the Néel temperature ($T_N = 132.2$ K) the magnetic system of the hexagonal close packing structure of Ho orders in the spin helix. Similar to Dy, the moments in Ho are ferromagnetically coupled within the basal (*ab*) plane, but their orientation rotates at a certain angle while moving along the *c* axis. It was, however, shown that, contrary to Dy, the magnetic order in bulk Ho is strongly affected by the crystal-field anisotropy [6,14,15]. The magnetic moments are bunching along the six easy axes in the basal plane due to the crystal-field anisotropy, which leads, first, to the lock-in of the wave vector into values commensurable with the atomic lattice in certain temperature intervals, and, second, to the formation of a series of long period commensurate spin-slip structures [6,14,15]. Particularly, the moments are ordered below 18 K in a commensurate cone structure with the wave vector **k** along the *c* axis, forming a 12-layer magnetic unit cell.

As it was shown in Ref. [6], the magnetic structure of Ho/Y multilayers is similar to that of bulk Ho. The coherent spin helix penetrates through the paramagnetic Y layers due to the charge density wave of the conduction electrons [1]. The effective turn angle in Y is found to be constant (about 51°) at all temperatures, while the turn angle in Ho layers was larger in comparison with bulk Ho. In addition, the ferromagnetic transition at 18 K is suppressed in multilayers. The strains introduced by the lattice mismatch between Ho and Y produce a lattice pressure which reduces the ordering temperature inside the Ho blocks. The corresponding lattice parameter in the bulk is equal to $c = 2.808$ Å for Ho and $c = 2.865$ Å for Y, respectively.

In this paper we show that the chiral symmetry of the degeneracy of the helix structure can be lifted by an in-plane magnetic field applied upon cooling of the holmium/yttrium multilayers. The effect of an applied magnetic field was studied using the following three samples: [Ho 45 Å/Y 30 Å]$_n$, [Ho 25 Å/Y 20 Å]$_n$ and [Ho 20 Å/Y 30 Å]$_n$, denoted as (Ho45Y30), (Ho25Y20), and (Ho20Y30), respectively. The number of bilayers n is 20 in (Ho45Y30) and (Ho25Y20), and 30 for (Ho20Y30). The samples were grown along the *c* axis [001] of the Ho and Y hcp structure by molecular-beam-epitaxy techniques at Uppsala University [16] on a sapphire substrate with a 150 Å Nb buffer layer and a 200 Å Y seed layer below the superlattice. The samples were capped by a 50 Å

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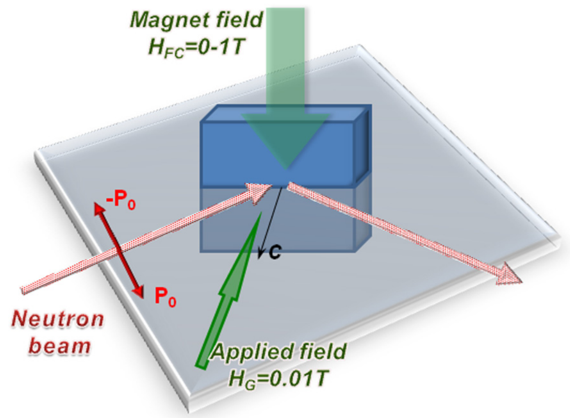

FIG. 1. (Color online) The schematic drawing of the experiment.

Nb layer to prevent the oxidation of the magnetic material. The good chemical and crystallographic quality of the SLs was verified by x-ray diffractometry and reflectometry using a standard x-ray diffractometer at the Helmholtz-Zentrum Geesthacht. The structural coherence lengths along the c axis are estimated from the full width at half maximum (FWHM) of the central Bragg peak and are between 450 and 650 Å with an average mosaicity of about 0.34°. The lattice parameters along the c axis are larger for the holmium blocks and smaller for yttrium blocks compared to the individual bulk materials.

To answer the question whether any preferable chirality arises for these structures, polarized neutrons are especially useful since they allow one to determine the chirality of magnetic structures [17]. The total magnetic elastic scattering cross section for polarized neutrons can be separated into a polarization-dependent contribution and a polarization-independent part. The latter part is also asymmetric with respect to the momentum transfer Q and can be associated with the average chirality of the magnetic system.

II. EXPERIMENTAL

The polarized neutron experiments were carried out at the MARIA reflectometer at the FRM II (JCNS). An incident neutron beam with a polarization $P = 0.98$, wavelength $\lambda = 6$ Å, and $\Delta\lambda/\lambda = 0.1$ were used. In order to provide a perpendicular guide field with respect to the sample plane at the sample position, additional magnetic guide fields were mounted. Due to the nontrivial setup, the polarization at the sample position was reduced to about $P_0 = 0.90$. The c axis of the multilayer sample was set perpendicular to the incident beam (Fig. 1). A magnetic field of up to 1 T could be applied parallel to the multilayer surface during the field cooling (FC) procedure from $T > T_N$ to $T < T_N$. The reflectivity profile at the Bragg peak position of the helices was taken at different temperatures after zero field cooling (ZFC) and the FC procedures from $T > T_N$ to $T < T_N$. The scattering intensity was measured in a small guide field H_G, with $(\mathbf{H}_G \| \mathbf{P}_0 \| \mathbf{k})$, and thereupon the in-plane field H_{FC} was switched off. The sense of the polarization followed a magnetic guide field of 1 mT applied perpendicularly to the multilayer surface (along the c axis). Such geometry was used to study the polarization-dependent part of the scattering cross section. At this configuration, with the polarization of the incident beam aligned along the direction of the applied field $(\mathbf{P}_0 \| \mathbf{Q})$, the corresponding scattered intensities $[I^+ = I(\mathbf{Q}, +\mathbf{P}_0)$ and $I^- = I(\mathbf{Q}, -\mathbf{P}_0)]$ are due to scattering on either the right- or left-handed domains, respectively. The average chirality, which is proportional to the difference in the population of the left- and right-handed helices, was measured as the polarization-dependent asymmetric part of the magnetic neutron-scattering cross section [10].

Thus we introduce here a chiral parameter directly related to the measured intensities and to the imbalance between the left- and right-handed domains:

$$\gamma = \left| \frac{1}{P_0} \frac{I(+P_0) - I(-P_0)}{I(+P_0) + I(-P_0)} \right|. \tag{1}$$

The measured value of γ was normalized to the polarization P_0 at the sample position.

III. RESULTS

Figure 2 shows reflectivity profiles, $I(+P_0)$ and $I(-P_0)$, for the sample Ho25Y20 after the ZFC procedure [Fig. 2(a)] and

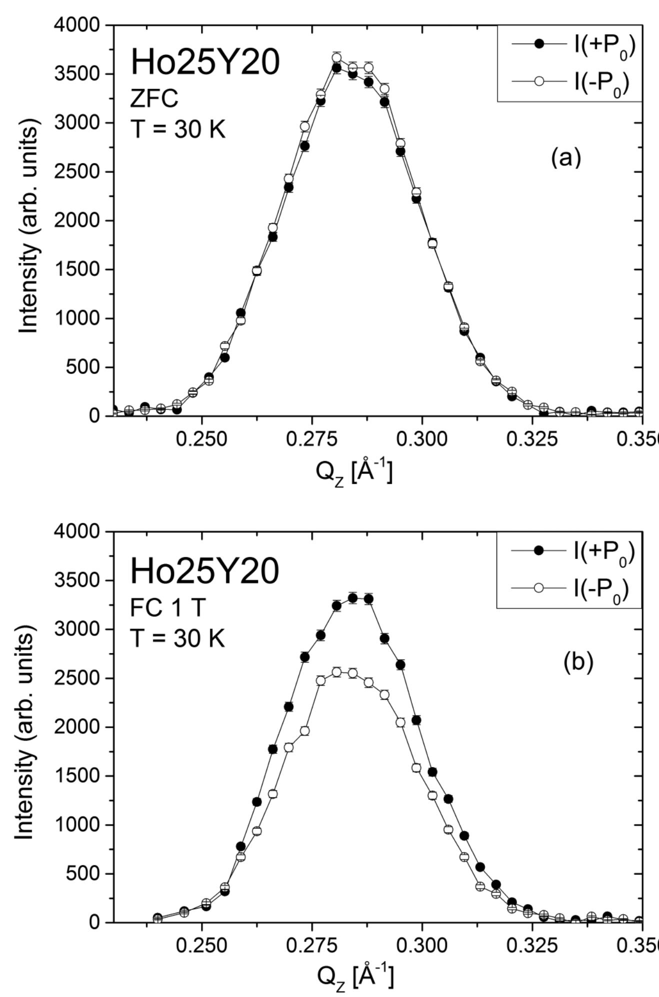

FIG. 2. The Q dependence of the neutron-scattering intensity (reflectivity profile) for the samples Ho25/Y20, taken for two polarizations of the incident beam at $T = 30$ K after the ZFC procedure (a) and FC procedure at an applied field $H = 1$ T (b).

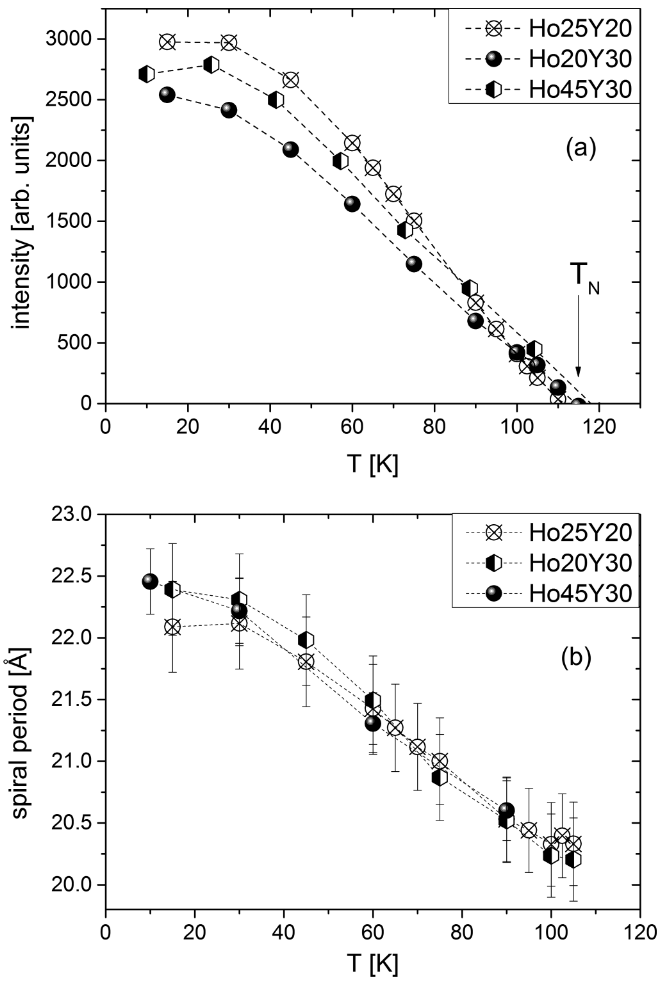

FIG. 3. Temperature dependence of magnetic intensity (a) and the spiral period in the Ho layer (b).

the FC procedure at $H = 1$ T cooled down to $T = 30$ K. The observed peaks obviously originate from the incommensurate helical spin structure since they appear only below T_N and at a Q value not corresponding to the structural superlattice period or a magnetically commensurate Q state. No difference in the scattering profiles is observed upon the ZFC procedure within the error bars [Fig. 2(a)]. The FC procedure, on the other hand, show a nonzero difference between the two scattering intensities of opposite polarizations, $I(+P_0)$ and $I(-P_0)$, demonstrating the appearance of a nonzero average chirality in the sample.

The temperature dependence of the integrated magnetic peak intensity after the FC procedure is shown in Fig. 3(a). We extrapolated the intensity of the magnetic peak to the zero value and found that the so determined ordering temperature T_N in Ho layers of these samples is significantly reduced with respect to the bulk material to 115 ± 3 K. It should be noted that the previously applied magnetic field H_{FC} does not affect the position of the magnetic peak and the spiral period does not depend on the applied field procedure. The spiral period d_s can be calculated from the peak center of magnetic reflection (Q_{cen})

$$d_s = \frac{2\pi}{Q_{\mathrm{cen}}}. \tag{2}$$

Figure 3(b) shows that the values of d_s are practically the same for the investigated samples and independent of the holmium and yttrium thicknesses of the individual sample. The spiral period d_s is equal to 22.3 ± 0.5 Å at low temperatures and decreases with increasing temperature to about 20 Å in the vicinity of T_N. As it was shown in Ref. [6], the scattering data can be reasonably modeled if one assumes that the phase shift across the Y layer, associated with a wave vector $\mathbf{k}_{\mathrm{Y}}$, is different from the wave vector of Ho ($\mathbf{k}_{\mathrm{Y}} \neq \mathbf{k}_{\mathrm{Ho}}$). According to Ref. [6], the value of $\mathbf{k}_{\mathrm{Y}}$ of 0.31 Å^{-1} is temperature independent, corresponding to a turn angle between the Y atomic planes along a c axis of 51° and a period of the helix of 20 Å. Thus we associate the changes of the spiral period d_s to changes in the phase shift across the Ho layer and to the changes of its wave vector $\mathbf{k}_{\mathrm{Ho}}$ with temperature.

Figure 4 shows the value of γ for the samples Ho25Y20, Ho20Y30, and Ho45Y30 in dependence of the temperature after the ZFC and FC procedures, respectively. After the ZFC procedure shown in Fig. 4(a), the chirality γ is close to zero within the error bars over the complete temperature range for all three samples. This observation can be easily understood

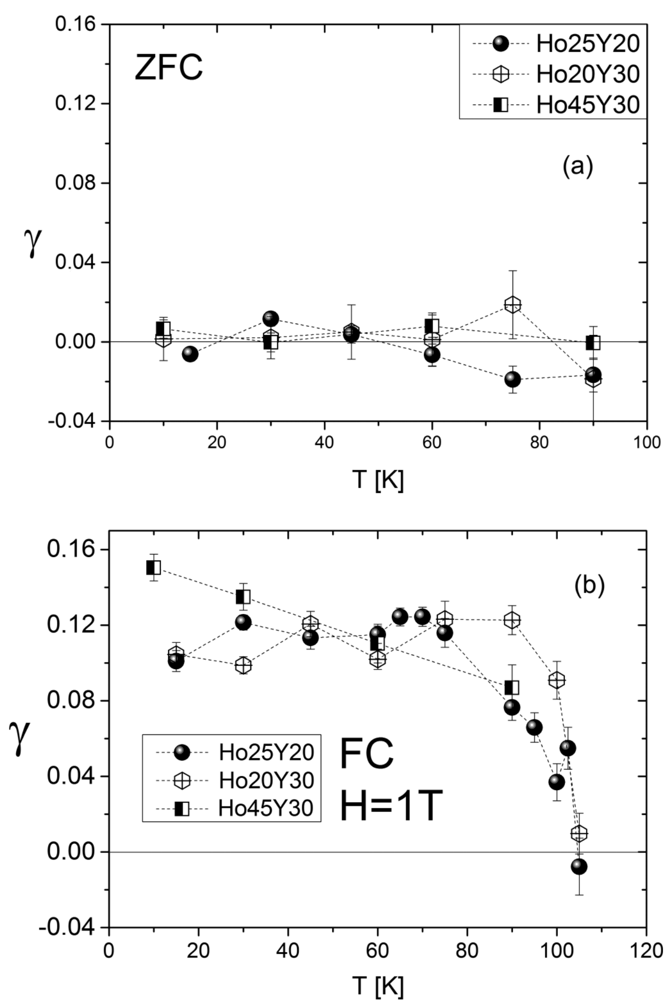

FIG. 4. The chirality for the samples Ho25Y20, Ho20Y30, and Ho45Y30 in dependence on temperature prehistory: (a) T dependence after ZFC, and (b) T dependence after FC under an applied field $H = 1$ T.

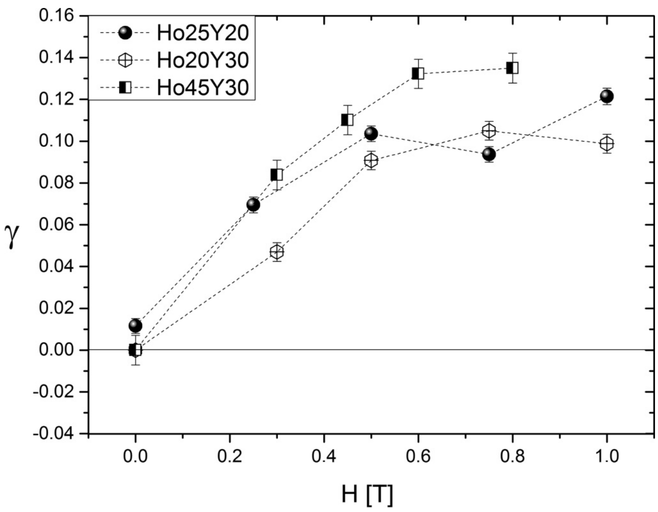

FIG. 5. The chirality for the samples Ho25Y20, Ho20Y30, and Ho45Y30 in dependence on field prehistory: H dependence after FC to 30 K.

considering the RKKY interaction as the dominant interaction for forming the helical structures [13]. In this case, the right- and left-handed helices are energetically equivalent to each other and both states will be occupied in equal measures. The value of chirality γ measured after the FC procedure in a magnetic field of 1 T, on the other hand, clearly shows nonzero values of up to 12%, suggesting strongly that the degeneracy of the chiral symmetry is now lifted. The value of γ drops sharply to zero when the temperature approaches T_N, indicating that the introduction of the chirality in the systems predominantly occurs in a very limited temperature range close to the transition temperature for all three samples. In Fig. 5 the chirality γ is plotted as a function of the applied field H_{FC} during the FC procedure with subsequent cooling down to a temperature of 30 K. The value of γ increases with the increase of the applied field in the range $H_{FC} < 0.5$ T, indicating that the strength of the field has a considerable influence on the magnetic structure of the samples in this regime. The chirality γ, however, saturates at a value of 0.10–0.12, with no further significant increase for $0.5 < H_{FC} < 1$ T. The FC procedure clearly demonstrates the predominance of one type of helix domain over the other one induced by the in-plane magnetic field applied during the FC procedure.

IV. DISCUSSION AND CONCLUSION

As it was already mentioned above, a very similar behavior with respect to the net chirality was detected earlier in Dy/Y multilayers. The assumption of a lifting of degeneracy of the chiral symmetry due to the appearance of a DMI on interfaces was theoretically confirmed by Haraldsen and Fishman in 2010 [18]. They had shown that interfacial defects, emerging due to the overlap between magnetic Dy and nonmagnetic Y atoms, can produce a nonzero DM contribution normal to the interface in magnetic heterostructures. Moreover, the appearance of a DMI was predicted on the perfect interfaces in Ref. [19]. Xia *et al.* have studied the interlayer exchange coupling in ferromagnetic-nonmagnetic multilayer structures, based on an extended Anderson s-d mixing model, and showed the rise in both the noncollinear DMI as well as the isotropic RKKY interaction between the neighboring ferromagnetic layers. They have showed that DM-type interlayer coupling oscillates with spacer layer thickness and has the same period as that of the usual RKKY term at a large spacer layer thickness, but with a phase shift of $\pi/2$. The value of DMI reaches 10% of the RKKY interaction.

This theoretical calculation, however, does not explain the link between DMI and an applied magnetic field. A possible interpretation is as follows: By the application of an in-plane magnetic field, particularly in the vicinity of the transition temperature where the RKKY interaction is weak and comparable to the applied Zeemann energy, the DMI can be coupled into the system and further transferred by the increasing strength of the RKKY interaction with decreasing temperature throughout the whole SL. Once the RKKY is strong enough, the imprinted chirality during the transition temperature remains unchanged.

We assume that the net chirality in Ho/Y systems has the same nature and appears due to the linking of the degeneracy of the symmetry at the magnetic-nonmagnetic interfaces. The x-ray characterizations clearly indicate that the interfaces of all of the examined Ho/Y SLs are of good quality, but are also partly intermixed and thus fulfill the necessary conditions for the occurrence of a nonzero DM energy contribution at the interfaces. On the other hand, the chirality of the investigated samples does not exceed a maximum value of about 13%, which is twice as less as that in Dy/Y multilayers [10,11]. We explain this decrease by a strong influence of the crystal-field anisotropy in Ho layers. In general, we have given experimental evidence for the Dzyaloshinskii-Moriya interaction on the interfaces of the magnetic multilayer structure taking Ho/Y multilayers, for example. Further experimental and theoretical studies can reveal the role of crystal-field anisotropy in Ho layers as well as the role of the possible interplay between the RKKY and DM interactions in these types of magnetic multilayer systems.

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[1] R. W. Erwin, J. J. Rhyne, M. B. Salamon, J. Borchers, S. Sinha, R. Du, J. E. Cunningham, and C. P. Flynn, Phys. Rev. B **35**, 6808 (1987).

[2] J. A. Borchers, M. B. Salamon, R. W. Erwin, J. J. Rhyne, R. R. Du, and C. P. Flynn, Phys. Rev. B **43**, 3123 (1991).

[3] C. de la Fuente, R. A. Cowley, J. P. Goff, R. C. C. Ward, M. R. Wells, and D. F. McMorrow, J. Phys.: Condens. Matter **11**, 6529 (1999).

[4] M. B. Salamon, S. Sinha, J. J. Rhyne, J. E. Cunningham, R. W. Erwin, J. Borchers, and C. P. Flynn, Phys. Rev. Lett. **56**, 259 (1986).

[5] R. A. Cowley, D. F. McMorrow, A. Simpson, D. Jehan, P. Swaddling *et al.*, J. Appl. Phys. **76**, 6274 (1994).

[6] D. A. Jehan, D. F. McMorrow, R. A. Cowley, R. C. C. Ward, M. R. Wells, N. Hagmann, and K. N. Clausen, Phys. Rev. B **48**, 5594 (1993).

[7] D. Haskel, G. Srajer, J. C. Lang, J. Pollmann, C. S. Nelson, J. S. Jiang, and S. D. Bader, Phys. Rev. Lett. **87**, 207201 (2001).

[8] Y. Choi, D. Haskel, A. Cady, J. C. Lang, D. R. Lee, G. Srajer, J. S. Jiang, and S. D. Bader, Phys. Rev. B **73**, 174401 (2006).

[9] S. V. Grigoriev, A. I. Okorokov, Yu. O. Chetverikov, D. Yu. Chernyshev, H. Eckerlebe, K. Pranzas, and A. Schreyer, JETP Lett. **83**, 478 (2006).

[10] S. V. Grigoriev, Yu. O. Chetverikov, D. Lott, and A. Schreyer, Phys. Rev. Lett. **100**, 197203 (2008).

[11] S. V. Grigoriev, D. Lott, Yu. O. Chetverikov, A. T. D. Grunwald, R. C. C. Ward, and A. Schreyer, Phys. Rev. B **82**, 195432 (2010).

[12] A. N. Bogdanov and U. K. Rossler, Phys. Rev. Lett. **87**, 037203 (2001).

[13] W. C. Koehler, J. W. Cable, M. K. Wilkinson, and E. O. Wollan, Phys. Rev. **151**, 414 (1966).

[14] D. A. Tindall, C. P. Adams, M. O. Steinitz, and T. M. Holden, J. Appl. Phys. **76**, 6229 (1994).

[15] J. Jensen, Phys. Rev. B **54**, 4021 (1996).

[16] C. F. Majkrzak *et al.*, Adv. Phys. **40**, 99 (1991).

[17] S. V. Maleyev, Phys. Rev. Lett. **75**, 4682 (1995).

[18] J. T. Haraldsen and R. S. Fishman, Phys. Rev. B **81**, 020404(R) (2010).

[19] K. Xia, W. Zhang, M. Lu, and H. Zhai, Phys. Rev. B **55**, 12561 (1997).