Effect of anisotropy on anomalous Hall effect in Tb–Fe thin films

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The electrical and Hall resistivities of Tb_xFe_{100-x} thin films in the temperature range 13–300 K were investigated. The sign of Hall resistivity at 300 K is found to change from positive for $x=28$ film to negative for $x=30$ film, in accordance with the compensation of Tb and Fe moments. All the films are seen to have planar magnetic anisotropy at 13 K. The temperature coefficients of electrical resistivities of the amorphous films with $19 \leq x \leq 51$ are seen to be negative. The temperature dependence of Hall resistivity of these films is explained on the basis of random magnetic anisotropy model. The temperature dependences of Hall resistivities of the $x=22$ and 41 films are seen to exhibit a nonmonotonous behavior due to change in anisotropy from perpendicular to planar. The same behavior is considered for the explanation regarding the probable formation of Berry phase curvature in these films. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3138807]

I. INTRODUCTION

Hall resistivity (HR) in a ferromagnetic material is a combination of both ordinary and anomalous Hall resistivities (OHR and AHR). The HR is given by $(\rho_H)_{mag} = \mu_0[R_0H + R_AM(H)] = \rho_{OHR} + \rho_{AHR}$, where R_0 and R_A are ordinary and anomalous Hall coefficients, respectively, and M is the magnetization.[1] Generally, the AHR is very large in magnitude compared to the OHR. Early interpretations of the anomalous Hall effect (AHE) have explained the qualitative features observed in experiments, including linear (skew scattering)[2] or quadratic (side jump),[3] in terms of longitudinal resistivity (ρ), based on asymmetric scattering of the spin polarized charge carriers in the presence of spin-orbit coupling. However, a quantitative agreement between the scattering theories and experiments remained largely unsettled, partly because the scattering potentials are unknown. Karplus and Luttinger[4] proposed that the spin-orbit coupling in Bloch bands gives rise to anomalous Hall conductivity (AHC) in ferromagnetic materials and these (scattering free) contributions have been rederived in a semiclassical work of wavepacket motion in Bloch bands by taking into account Berry phase effects.[5] The AHC has been obtained as an integral of the Berry curvature $\Omega_n(\boldsymbol{k})$ over the occupied electron states in $\boldsymbol{k}$-space and is given by[6,7]

$$\sigma_{xy} = -\frac{e^2}{\hbar}\int_{BZ}\frac{d^3\boldsymbol{k}}{(2\pi)^3}\Omega_n(\boldsymbol{k}).$$

Inspired by the new insight of the Berry phase effects on Bloch electrons, several groups have evaluated the intrinsic AHC for ferromagnetic semiconductors,[8] Fe,[6] Gd,[9] Mn_5Ge_3,[10] and oxides,[11–13] from the first-principles calculations. R-Fe (R is the rare earth element) thin films are known to exhibit perpendicular magnetic anisotropy (PMA) at room temperature, and several of these films find applications in magneto-optic recording devices.[14] The PMA can be observed from the perpendicular magnetization measurements as well as through the Hall effect measurements.[15] By virtue of their large and ordered magnetic moments, R-Fe thin films are also known to exhibit AHE.[16] Harris *et al.*[17] proposed "random magnetic anisotropy" (RMA) model in amorphous $TbFe_2$ alloys, according to which, each Tb moment is in a local anisotropy field of random orientation. The strength and orientation of local magnetic anisotropy of the Tb ion tend to cause the local magnetic moment to align in a direction away from a collinear arrangement.[18] The effect of RMA on AHE has not yet been known and therefore an attempt was made to investigate the effect of anisotropy (both RMA and PMA) on AHE. In the present work, we report the HR and electrical resistivity of Tb–Fe thin films in the temperature range 13–300 K.

II. EXPERIMENTAL DETAILS

The Tb_xFe_{100-x} ($x=9$, 19, 22, 28, 30, 41, and 51 at. %) thin films were prepared by cosputtering Tb and Fe employing a dc magnetron sputtering system. The films were deposited on thermally oxidized Si substrates at a pressure of 2.5×10^{-3} mbar. The base pressure of the chamber was 3×10^{-6} mbar and argon was used as the sputtering gas. The deposition rates and thicknesses of the films were determined by an *in situ* quartz crystal thickness monitor and the average deposition rate was about 2.8 Å/s. The films were prepared by varying the deposition rates of Tb (0.6–2.4 Å/s) and Fe (0.4–2.1 Å/s) and were about 200 nm thick. X-ray diffraction (XRD) patterns of the films were taken using a PANalytical (X'pert PRO) x-ray diffractometer employing Cu $K\alpha$ radiation. Compositional analyses of films were carried out using energy dispersive analysis of x rays in a scanning electron microscope (FEI, Quanta-200). Atomic force microscopy (AFM) measurements were carried out employing Nanoscope IV-Dimension 3100 SPM system. HR and electrical resistivity were measured by van der Pauw method in the

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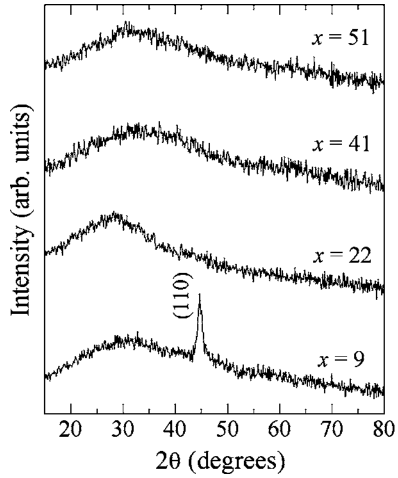

FIG. 1. XRD patterns of Tb_xFe_{100-x} (x=19, 22, 41, and 51) thin films.

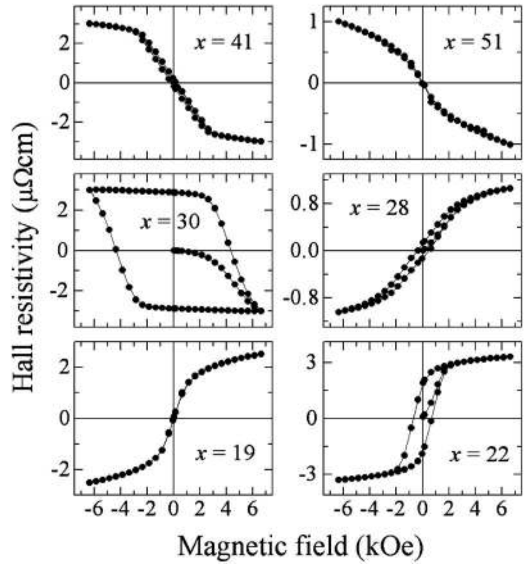

FIG. 3. HR of Tb_xFe_{100-x} films at room temperature.

temperature range 13–300 K. The HR measurements were carried out on films of square geometry of 6 mm side in magnetic fields up to ±6.5 kOe, with the magnetic field applied perpendicular to the plane of the film.

III. RESULTS AND DISCUSSION

The XRD patterns reveal that the as grown Tb_xFe_{100-x} ($19 \leq x \leq 51$) films are amorphous, as shown in Fig. 1. The x=9 film is found to contain α-Fe phase along with the amorphous Tb–Fe phase. The AFM images of x=9 and x=41 films are shown in Fig. 2. The crystallite size is seen to vary from 60 to 90 nm for x=9 film in the above image, and the average crystallite size from the Scherrer formula[19] is found to be 84 nm. The x=41 film was found to have formed in amorphous phase as observed through the XRD pattern, and the amorphous nature is clearly observed through the AFM image where diffused crystallites of ≤10 nm are seen.

In order to discuss the effect of anisotropy on the transport properties, results are reported only for the amorphous films and the x=9 film is not included in further discussions. The results of HR and electrical resistivity measurements in the temperature range 13–300 K are discussed in Secs. III A–III D.

A. HR at room temperature (300 K)

The Hall resistivities of Tb_xFe_{100-x} (x=19, 22, 28, 30, 41, and 51 at. %) thin films are shown in Fig. 3. The Hall resistivities determined with magnetic fields +H and −H are denoted by ρ_{H+} and ρ_{H-}, respectively. The ρ_{H+} of Tb_xFe_{1-x} films with $x \leq 28$ are seen to be positive, whereas those of the films with $x \geq 30$ are seen to be negative. This result is in

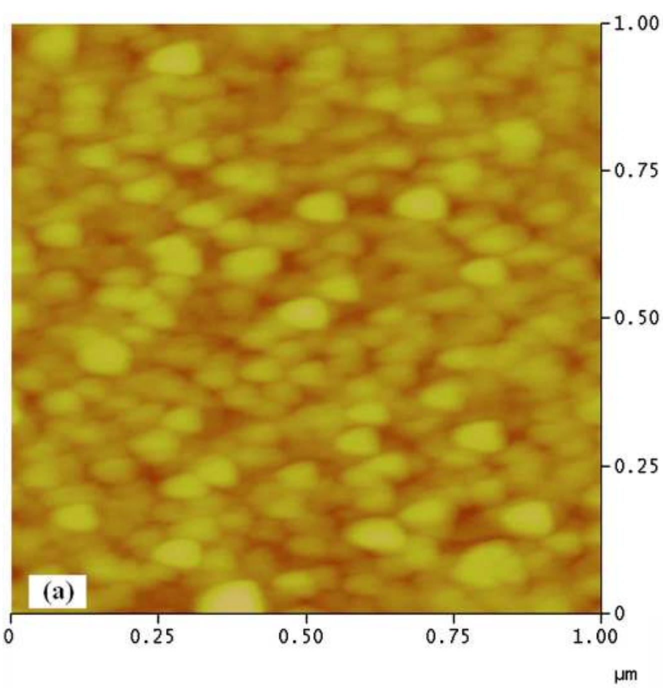

FIG. 2. (Color online) AFM images of Tb_xFe_{100-x} films of (a) x=9 and (b) x=41 contents.

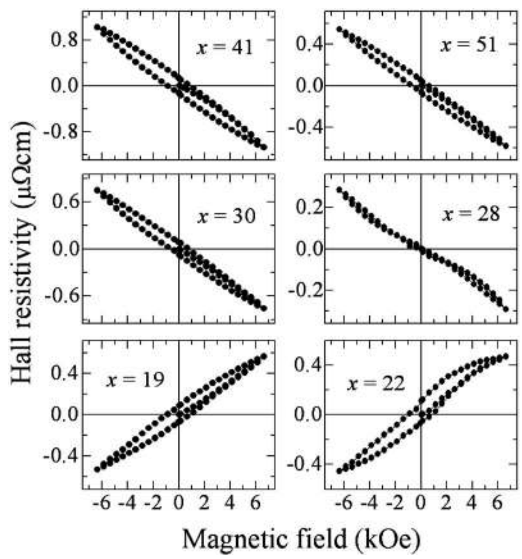

FIG. 4. HR of Tb_xFe_{100-x} films at 13 K.

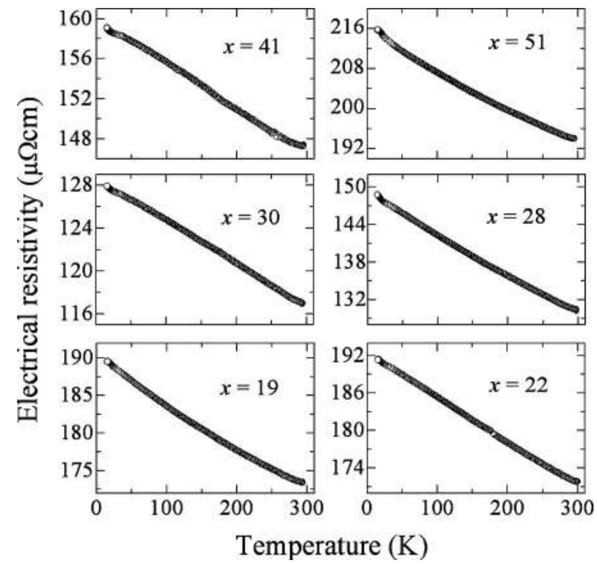

FIG. 5. Electrical resistivity of Tb_xFe_{100-x} films in the temperature range 13–300 K.

accordance with the fact that in Tb–Fe films, Tb and Fe moments are antiparallel to each other[20] and that the moment of Fe is dominant in the Fe-rich case and that of Tb is dominant in the Tb-rich case.

The HR data of the $x=19$ film indicate that there is a tendency toward PMA in this film. On the other hand, the Hall resistivities of films with $x=22$ and 41 reveal that there is PMA in these films. In spite of the effect of crystalline electric fields being small in amorphous materials, the presence of PMA is mainly due to a significant spin-orbit coupling. This indicates that the anisotropy is single ion like in these films. PMA in *R-TM* (TM is the transition metal) films has been proposed based on the hypothesis that during the layer-by-layer growth, small planar hexagonal units formed, defining a net direction that is perpendicular to the film plane.[21] Harris *et al.*[22] reported that the PMA in amorphous Tb–Fe films is associated with larger Tb–Fe correlations perpendicular to the film than in the plane of the film. The saturation magnetization of the $Tb_{28}Fe_{72}$ film has been reported to be small due to the near compensation of the Tb and Fe moments.[23] In the present investigation, very close to the compensation concentration (between 28 and 30 at. %), as anisotropy is very small, the demagnetizing field tends to keep the moments in the plane of the film. This is reflected in the $x=28$ film where significant planar anisotropy is seen to ride over the PMA. The moments in the $x=30$ film are expected to lie almost in the plane of the film, as seen through the large hysteresis and knee field at above 6 kOe. The $x=51$ film is seen to have planar anisotropy.

B. HR at 13 K

The Hall resistivities of Tb_xFe_{100-x} films $(19 \leq x \leq 51)$ were measured at 13 K after the films were cooled in zero magnetic field [zero field cooled (ZFC) measurements]. PMA is not observed in any of the films at 13 K, as shown in Fig. 4, though some are seen to exhibit PMA at 300 K. Though the strength of spin-orbit coupling is large at low temperatures, spin reorientation transitions have not been observed in either bulk or thin films of $TbFe_2$.[24] Therefore, in the present investigation, the absence of PMA at low temperatures could be due to increase in strain induced anisotropy (planar) because of large magnetostriction. The Hall resistivities are found to be small at 13 K compared to those at 300 K (Fig. 3), in accordance with the proposal of the absence of PMA in these films. The $x=28$ film is seen to have negative ρ_{H+} at 13 K due to the dominant Tb moment and positive ρ_{H+} at 300 K (Fig. 3) due to the dominant Fe moment.

C. Electrical resistivity

The electrical resistivity data of Tb_xFe_{100-x} (19, 22, 28, 30, 41, and 51 at. %) thin films in the temperature range 13–300 K are shown in Fig. 5. The electrical resistivities of all the films in the concentration region discussed are seen to increase upon decrease in temperature. The temperature coefficient of resistivity in amorphous materials has been calculated from Ziman's[25] theory proposed in liquid transition metals based on the structure factor. The phonons and thermal variations in the structure factor can give rise to positive and negative temperature coefficients.[25,26] In the present investigation, PMA (or a tendency for PMA) was observed in the Tb_xFe_{100-x} films with $19 \leq x \leq 51$ content through HR at 300 K, and all the films were seen to be free from PMA at 13 K. This may be due to the larger density of the 4*f* electrons within the plane, led by strong spin-orbit interactions. As the films are amorphous, within the plane of the film, the anisotropy in the resistivity could have resulted from the additional scattering of electrons by the 4*f* quadrupole moments.[27]

D. Temperature dependence of Hall resistivity

Figure 6 shows the HR (ρ_{H+}) at 6 kOe of Tb_xFe_{100-x} (19, 22, 28, 30, 41, and 51 at. %) thin films in the temperature range 13–300 K. The measurements were carried out while cooling the films in the presence of the magnetic field [field

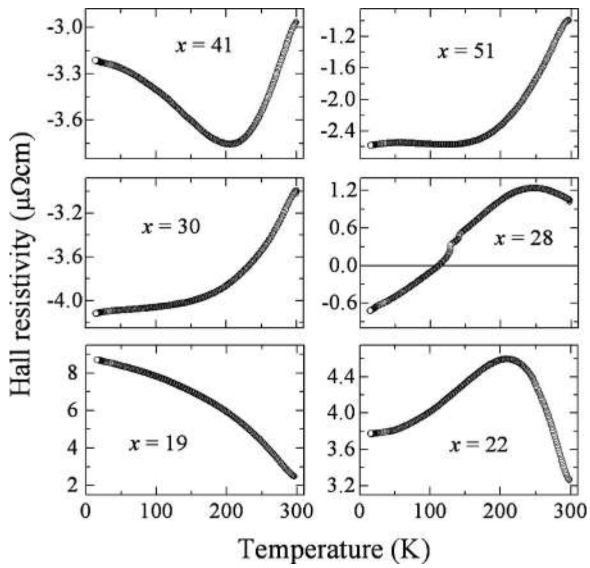

FIG. 6. HR of Tb_xFe_{100-x} films in the temperature range 13–300 K.

cooled (FC) measurements]. The magnitudes of HRs of $x=19$ and 51 films are seen to increase with decreasing temperature and this can be explained by RMA model. According to RMA model, when an amorphous ferromagnetic thin film is cooled in the presence of a magnetic field (FC), then at low temperatures, the magnetic moments lie in a direction decided by competition between the anisotropy energy (local crystalline electric field) and exchange energy (aided by the external magnetic field).[17,28] In the present investigation, the magnetic moments in $x=19$ and 51 films are expected to lie more or less in the plane of the film with presumption that the (planar) anisotropy does not change much with temperature since there is no PMA (except a weak tendency) in these films. Thus, as the temperature decreases, due to the increase in net moment because of domination of the Tb moment, the HR increases. When the amorphous films are ZFC, the magnetic moments contain a correlated speromagnetic state (metastable disordered state)[29] which has small net moment that cannot be reoriented by the order of fields applied in the present investigation (±6.5 kOe), and this explains the low HR values at 13 K.

The magnitudes of HRs of $x=22$ and 41 films are seen to increase with decreasing temperature and reach respective maximum values at 209 and 206 K, respectively, before decreasing at lower temperatures. The increase in HR is in accordance with increasing net moment (as per the discussion of $x=19$ and 51 films) and the decrease in HR at low temperatures could be attributed to the change in anisotropy from perpendicular to planar (moments freeze in a direction decided by local anisotropy). The magnitude of HR (HR is negative) of the $x=30$ film is seen to increase with decreasing temperature. Since it has a large anisotropy, the HR of the above film is expected to follow those of the $x=19$ and 51 films. However, the HR values of $19 \leq x \leq 51$ films in ZFC (at 13 K and 6 kOe) are seen to be small compared to that from the FC measurements, in accordance with the RMA model.

In the recent investigations, the nonmonotonous variation in HR in crystalline magnetic materials with temperature has been related to the appearance of Berry phase.[10–13] The AHC was expressed as an integral of the Berry curvature over occupied electron states in $\boldsymbol{k}$-space and it has been shown that the variation in HR with either temperature or magnetic field (with respect to knee profiles) is nonmonotonous.[11,12] Taguchi *et al.*[12] reported that the temperature and field dependence of AHC is nonmonotonous within the conventional view of AHE by an observation that the relevant variable is the directional degree of freedom, rather than the magnitude, of the spin dependent magnetic moment. However, the present investigation is limited to amorphous materials and thus, the temperature variation in HR may not be a direct manifestation of Berry phase though the dependence on the local directional freedom could have some influence. The nonmonotonous behavior of HR with respect to temperature in $x=22$ and 41 films seems to be comparable to the behavior usually observed in the formation of Berry phase curvature in accordance with the proposed noncoplanar spin configuration due to the anisotropy.

IV. SUMMARY AND CONCLUSIONS

The sign of HR at 300 K is found to change (associated with a change in anisotropy) from positive for $x=28$ to negative for $x=30$ content in Tb_xFe_{100-x} thin films, in accordance with the compensation of Tb and Fe moments. All the films are seen to have planar anisotropy at 13 K. The films with $19 \leq x \leq 51$ content are seen to exhibit negative temperature coefficient of electrical resistivity in the temperature range 13–300 K in accordance with the development of in-plane anisotropy at low temperatures. The HRs of the above films are explained based on the RMA model with a significant contribution of change in anisotropy from perpendicular (PMA) to planar (for $x=22$ and 41 films). The $x=22$ and 41 films are seen to have a nonmonotonous behavior of HR with respect to temperature, and the same behavior is considered for the explanation regarding the probable formation of Berry phase curvature in these films.

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[1]C. L. Chien and C. R. Westgate, *The Hall Effect and Its Applications* (Plenum, New York, 1979).
[2]J. Smit, Physica (Utrecht) **21**, 877 (1955); **24**, 39 (1958).
[3]L. Berger, Phys. Rev. B **2**, 4559 (1970).
[4]R. Karplus and J. M. Luttinger, Phys. Rev. **95**, 1154 (1954).
[5]G. Sundaram and Q. Niu, Phys. Rev. B **59**, 14915 (1999).
[6]Y. Yao, L. Kleinman, A. H. MacDonald, J. Sinova, T. Jungwirth, D. S. Wang, E. Wang, and Q. Niu, Phys. Rev. Lett. **92**, 037204 (2004).
[7]D. Xiao, J. Shi, and Q. Niu, Phys. Rev. Lett. **95**, 137204 (2005).
[8]T. Jungwirth, Q. Niu, and A. H. MacDonald, Phys. Rev. Lett. **88**, 207208 (2002).
[9]S. A. Baily and M. B. Salamon, Phys. Rev. B **71**, 104407 (2005).
[10]C. Zeng, Y. Yao, Q. Niu, and H. H. Weitering, Phys. Rev. Lett. **96**, 037204 (2006).
[11]Z. Fang, N. Nagaosa, K. S. Takahashi, A. Asamitsu, R. Mathieu, T. Ogasawara, H. Yamada, M. Kawasaki, Y. Tokura, and K. Terakura, Science **302**, 92 (2003).
[12]Y. Taguchi, Y. Oohara, H. Yoshizawa, N. Nagaosa, and Y. Tokura, Science

291, 2573 (2001).
[13]R. Mathieu, A. Asamitsu, H. Yamada, K. S. Takahashi, M. Kawasaki, Z. Fang, N. Nagaosa, and Y. Tokura, Phys. Rev. Lett. **93**, 016602 (2004).
[14]P. Hansen and H. Heitmann, IEEE Trans. Magn. **25**, 4390 (1989); P. Hansen, C. Clausen, G. Much, M. Rosenkranz, and K. Witter, J. Appl. Phys. **66**, 756 (1989).
[15]T. W. Kim, S. H. Lim, and R. J. Gambino, J. Appl. Phys. **89**, 7212 (2001).
[16]R. Malmhäll, J. Appl. Phys. **54**, 5128 (1983).
[17]R. Harris, M. Plischke, and M. J. Zuckermann, Phys. Rev. Lett. **31**, 160 (1973).
[18]F. Hellman, A. L. Shapiro, E. N. Abarra, R. A. Robinson, R. P. Hjelm, P. A. Seeger, J. J. Rhyne, and J. I. Suzuki, Phys. Rev. B **59**, 11408 (1999).
[19]B. D. Cullity, *Elements of X-ray Diffraction*, 2nd ed. (Addison-Wesley, New York, 1978).
[20]J. J. Rhyne, S. J. Pickart, and H. A. Alperin, Phys. Rev. Lett. **29**, 1562 (1972).
[21]D. Mergel, H. Heitmann, and P. Hansen, Phys. Rev. B **47**, 882 (1993).
[22]V. G. Harris, K. D. Aylesworth, B. N. Das, W. T. Elam, and N. C. Koon, Phys. Rev. Lett. **69**, 1939 (1992).
[23]J. Huang, C. Prados, J. E. Evetts, and A. Hernando, Phys. Rev. B **51**, 297 (1995).
[24]U. Atzmony and M. P. Dariel, Phys. Rev. B **13**, 4006 (1976); A. Mougin, C. Dufour, K. Dumesnil, and Ph. Mangin, *ibid.* **62**, 9517 (2000).
[25]J. M. Ziman, Proc. R. Soc. London, Ser. A **318**, 401 (1970); R. Evans, D. A. Greenwood, and P. Lloyd, Phys. Lett. **35A**, 57 (1971).
[26]S. R. Nagel, Phys. Rev. B **16**, 1694 (1977).
[27]A. Fert and R. Asomoza, J. Appl. Phys. **50**, 1886 (1979).
[28]T. Kaneyoshi, *Introduction to Amorphous Magnets* (World Scientific, Singapore, 1992).
[29]J. M. Ruiz, X. X. Zhang, C. Ferrater, and J. Tejada, Phys. Rev. B **52**, 10202 (1995).