November 2004

Noncollinear spin states and competing interactions in half-metals

Ralph Skomski
University of Nebraska-Lincoln, rskomski2@unl.edu

Jian Zhou
University of Nebraska-Lincoln

Peter A. Dowben
University of Nebraska-Lincoln, pdowben@unl.edu

David J. Sellmyer
University of Nebraska-Lincoln, dsellmyer@unl.edu

Noncollinear spin states and competing interactions in half-metals and magnetic perovskites

R. Skomski,[a] J. Zhou, P. A. Dowben, and D. J. Sellmyer
Department of Physics and Astronomy and Center for Materials Research and Analysis, University of Nebraska, Lincoln, Nebraska 68588

(Presented on 8 November 2004; published online 4 May 2005)

The high-field spin structure of magnetic perovskites and related magnetoresistive materials is investigated by model calculations. Competing exchange as well as real-structure-dependent random field, random anisotropy, and Dzyaloshinskii–Moriya interactions yield a noncollinear magnetic structure that may be called a spin colloid. The noncollinear structure, which contributes to the zero- and finite-temperature spin mixing and reduces the magnetoresistance, is strongly field dependent. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1851412]

I. INTRODUCTION

The spatial magnetization distribution or spin structure is of crucial importance for the magnetoresistive behavior of perovskites and related materials, such as half-metallic ferromagnets. Typical experiments are performed in high magnetic fields, where the spin state is close to saturation and the magnetoresistive behavior approaches that of ideal half-metallics.[1,2] In low fields or at remanence, the same samples exhibit huge drops in band polarization, as observed for NiMnSb.[3,4] This deviation from perfect spin alignment results in spin mixing and negatively affects the magnetoresistance of half-metallic ferromagnets.[2,5]

The materials of interest tend to exhibit a subtle competition between ferromagnetic (FM) and antiferromagnetic (AFM) interactions, specifically in the vicinity of charge-ordering transitions.[6] In addition, in some half-metals there exist incommensurate spin states[2,7] of similar origin. The situation is further complicated by the simultaneous presence of random magnetocrystalline anisotropy and Dzyaloshinskii–Moriya (DM) interactions at the grain boundaries. Here we use a generalized nanomagnetic or "micromagnetic" approach to study the field dependence of the magnetization.

II. ENERGY CONTRIBUTIONS

The key to the understanding of noncollinear spin structures is the relativistic classification of the underlying mechanisms.[8,9] The starting point is the Pauli expansion of the Dirac interaction in terms of the electron velocity $v=\alpha c$, where $\alpha=4\pi\varepsilon_o e^2/\hbar c\approx 1/137$ is Sommerfeld's fine-structure constant.[10] For example, expanding the free-electron energy $mc^2\sqrt{1+v^2/c^2}$ yields the rest energy mc^2, the electrostatic energy $mv^2/2$, and the lowest-order relativistic correction $(\alpha/2)^2\, mv^2/2$. In the magnetic analogy, the last contribution describes, for example, magnetostatic interactions and magnetocrystalline anisotropy.

Subsequent terms in the expansion differ by factors of the order of $\alpha^2=1/137^2$. This is an order-of-magnitude estimate for magnetic and anisotropic forces, as compared to electrostatic forces. For example, typical susceptibilities are of the order of α^2 because they reflect the competition between magnetic and electrostatic interactions.

A. Heisenberg exchange

Heisenberg exchange is a relatively strong interaction of electrostatic origin and has the familiar structure $J(\mathbf{R}_i-\mathbf{R}_j)\,\mathbf{S}_i\mathbf{S}_j=J_{ij}\,\mathbf{S}_i\mathbf{S}_j$. It is *isotropic*, so that uniform spin rotation does not change the exchange energy, even if the system is noncubic. For example, layered structures tend to exhibit bond anisotropy, that is, intra- and interlayer exchanges may be different,[11] but the exchange does not depend on whether the magnetization is in plane or normal to the layers.

On a continuum level, the anisotropic exchange is described by

$$E_{\mathrm{ex}}=\int\int J(\mathbf{r}-\mathbf{r}')\mathbf{M}(\mathbf{r})\mathbf{M}(\mathbf{r}')dVdV', \tag{1}$$

as compared to the more familiar expression $\int A(\nabla\mathbf{M})^2dV$. Fourier transformation diagonalizes $J(\mathbf{r}-\mathbf{r}')$ and yields terms of the type $J(\mathbf{k})$, as compared to $A\mathbf{k}^2$. Noncollinear or incommensurate spin states then correspond to a minimum of $J(\mathbf{k})$. Examples are the helimagnetism of elemental rare-earth metals,[12] characterized by a k vector parallel to the c axis, and the spin structure of some semi-Heusler alloys, where the k vector $\mathbf{k}_o$ is given by the intercept of transverse-optical phonon and magnon modes.[2]

Note that the bond anisotropy described in Eq. (1) must not be confused with the proper exchange anisotropy involving exchange constants J_{xx}, J_{yy}, and J_{zz} rather than the isotropic constants J. The latter anisotropy is a small relativistic correction to the isotropic exchange and will be neglected.

B. Relativistic interactions

Second-order *magnetocrystalline anisotropy*, as exploited permanent magnets and magnetic recording media, is of the type $\Sigma_{\alpha\beta}\,K_{\alpha\beta}\,M_\alpha\,M_\beta$, where $K_{\alpha\beta}$ is a 3×3 spin-space

[a]Electronic mail: rskomski@unlserve.unl.edu

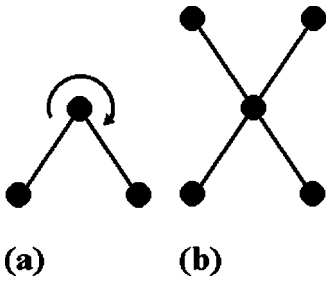

FIG. 1. Simple real-space illustration of Dzyaloshinskii–Moriya (DM) interactions: (a) nonzero interaction in magnets with low symmetry and (b) absence of DM interactions for crystals with inversion symmetry.

anisotropy tensor. Another relativistic contribution is the DM interaction

$$H_{\mathrm{DM}} = -\tfrac{1}{2}\Sigma_{ij}\mathbf{D}_{ij}\mathbf{S}_i\mathbf{S}_j. \tag{2}$$

In an itinerant description, the vector $\mathbf{D}_{ij}$ is proportional to $(\mathbf{R}_i-\mathbf{R}_o)(\mathbf{R}_j-\mathbf{R}_o)$, where $\mathbf{R}_o$ is the position of the not necessarily magnetic atom that mediates the interaction.[13] To be operative, the DM interaction requires local environments with sufficiently low symmetry (absence of inversion symmetry). It occurs, for example, in some crystalline materials, such as α-Fe_2O_3 (haematite), in amorphous magnets,[12] and in spin glasses.[8,12,13] They are also encountered in magnetic nanostructures,[14] where they have recently been discussed in a different context.[15]

Figure 1 illustrates the physical origin of the DM interaction. In solids with low symmetry, Fig. 1(a), the hopping electrons are able to benefit from the spin-orbit interaction in triangular configurations, thereby establishing a well-defined axial vector $\mathbf{D}_{ij}$ perpendicular to the plane of hopping. In crystals with inversion symmetry, Fig. 1(b), the corresponding DM contributions cancel by symmetry.[16] A major effect of the DM interaction is *spin canting*. Due to the relativistic nature of the DM interaction, typical canting angles are small, of the order of 0.1°, but they are easy to measure in structures whose underlying nonrelativistic spin structure is antiferromagnetic. (This yields the small net moment of so-called weak ferromagnets, such as α-Fe_2O_3.) The relative importance of DM interactions is particularly pronounced in the vicinity of FM-AFM and other exchange-related transitions.

The *magnetostatic self-interaction* field is of relativistic origin, too. In inhomogeneous magnets, it is obtained by Fourier transformation,[17] and for strong magnetic fields, the structure of the term is $\mathbf{k}\otimes\mathbf{k}/k^2$. Compared to exchange, $A\mathbf{k}^2$, magnetostatic interactions do not vanish for $k=0$, so that they are important on mesoscopic and especially macroscopic length scales.[9,14] Aside from the k dependence, the magnetostatic self-interaction term is very similar to the magnetocrystalline anisotropy, so that we incorporate it into $K_{\alpha\beta}$.

III. CALCULATION AND RESULTS

A. Basic formalism

For well-textured hard-magnetic materials,[8] the local magnetization $\mathbf{M}(\mathbf{r})$ is obtained by minimizing the micromagnetic energy functional[18]

$$\mathrm{E} = \int\left\{A(\mathbf{r})[\nabla(\mathbf{M}/M_s)]^2K(\mathbf{r})\frac{[\mathbf{n}(\mathbf{r})\mathbf{M}]^2}{M_s^2} - \mu_o\mathbf{M}\mathbf{H}\right\}dV. \tag{3}$$

Here $A(\mathbf{r})$ is the local exchange stiffness, $K_1(\mathbf{r})$ is the first uniaxial anisotropy constant, and $\mathbf{H}$ is the external magnetic field. Minimizing and linearizing Eq. (3) for a field $\mathbf{H}=H\,\mathbf{e}_z$ yields

$$-A\nabla^2\mathbf{m} + K_1\mathbf{m} + \frac{1}{2}\mu_oM_sH\mathbf{m} = K_1(\mathbf{r})\mathbf{a}(\mathbf{r}). \tag{4}$$

As explained elsewhere,[8,14] the derivation of this equation exploits $\mathbf{M}(\mathbf{r})=M_sm(\mathbf{r})^2\mathbf{e}_z/2+M_s\mathbf{m}(\mathbf{r})$ and $\mathbf{n}(\mathbf{r})=a(\mathbf{r})^2\mathbf{n}_o/2+\mathbf{a}(\mathbf{r})$.

Note that Eqs. (3) and (4) ignore the field dependence of the atomic magnetic moments, $M_s=M_s(H)$. In magnetoresistive oxides, this effect is not necessarily small, but the change is parallel to the external field and has no lowest-order effect on the relevant perpendicular magnetization component.

Equations (3) and (4) apply to simple ferromagnets but are easily generalized to the present materials. First, to describe competing exchange, the exchange-stiffness term in Eq. (4) must be replaced by an integral $\int J(\mathbf{r}-\mathbf{r}')\,\mathbf{m}(\mathbf{r}')dV'$. Second, both the source or force term $\mathbf{f}(\mathbf{r})=K_1(\mathbf{r})\,\mathbf{a}(\mathbf{r})$ and the "effective-anisotropy" term $K_1(\mathbf{r})+\mu_o\,M_s(\mathbf{r})\,H/2$ become more complicated. In particular, there is a random DM or spin-canting contribution to $\mathbf{f}(\mathbf{r})$.

Adding the contributions discussed in Sec. II yields the generalized linear micromagnetic equations

$$\int J(\mathbf{r}-\mathbf{r}')\mathbf{m}(\mathbf{r}')dV' + Q\mathbf{m} + \frac{1}{2}\mu_oM_sH\mathbf{m} = \mathbf{f}(\mathbf{r}), \tag{5}$$

where

$$Q = \begin{pmatrix} K_{zz}-K_{xx} & K_{xy} \\ K_{xy} & K_{zz}-K_{yy} \end{pmatrix} \tag{6a}$$

and

$$\mathbf{f} = K_{xz}\mathbf{e}_x + K_{yz}\mathbf{e}_y + \frac{1}{2}M_s^2\Sigma_j(D_{ij,y}\mathbf{e}_x - D_{ij,x}\mathbf{e}_y). \tag{6b}$$

Here the summation over j includes all neighbors of the atom at $\mathbf{r}_i$ that contribute to the DM interaction.

B. Approach to saturation

Equations (5) and (6) describe the high-field spin structure of the magnet. Assuming that H is large, we can solve Eq. (5) by series expansion. For strong fields, the leading interaction is of the type $J(\mathbf{r}-\mathbf{r}')+h\;\delta(\mathbf{r}-\mathbf{r}')/2$, where $h\sim H$. Since the addition of a unit operator, $\delta(\mathbf{r}-\mathbf{r}')$, does not change the eigenmodes of an operator, the noncollinearity of the magnetization state is given by the eigenmodes of $J(\mathbf{r}-\mathbf{r}')$. If the exchange was ferromagnetic, the eigenmodes would be plane waves (spin waves), and the spin structure would be ferromagnetic ($k=0$) with some random nanoscale

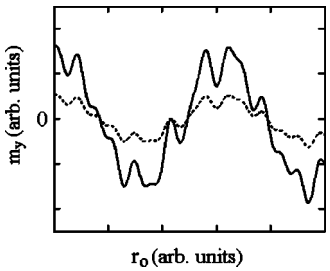

FIG. 2. Typical transverse magnetization modes (the spatial coordinate $\mathbf{r}_o$ is measured along $\mathbf{k}_o/\mathbf{k}_o$). Recall that $M_z(\mathbf{r})=M_s[1-m_x(\mathbf{r})^2-m_y(\mathbf{r})^2]^{1/2}$.

modulation due to random local anisotropy ($k>0$). In antiferromagnets, the presence of sublattices slightly complicates the structure of Eqs. (3) and (4).[19]

In general, the eigenmodes of $J(\mathbf{r}-\mathbf{r}')$ are incommensurate, that is, the wave vector $\mathbf{k}_o$ does not correspond to an integer multiple of the lattice constant. Figure 2 illustrates this point by showing the magnetization component m_y along $\mathbf{k}_o/k_o$ The main oscillation reflects the competing exchange, $J(\mathbf{r}-\mathbf{r}')$, whereas the noise is due to DM and other random contributions.

C. Two-mode description

In k space, the whole spectrum of k values is necessary to explain the spin structure of the system. A semiquantitative solution is to restrict the consideration to the most important wave vectors: $\mathbf{k}=0$ and $\mathbf{k}=\mathbf{k}_o$. Projecting the problem onto the two modes yields the equations

$$\langle \mathbf{m} \rangle = \frac{\langle \mathbf{f} \rangle}{h + \Delta J/2} \tag{7a}$$

and

$$\mathbf{m}_o = \frac{\mathbf{f}_o}{h - \Delta J/2}. \tag{7b}$$

Here $\langle \mathbf{m} \rangle$ and $\langle \mathbf{f} \rangle$ are the transverse-magnetization and random-force volume averages, $\mathbf{m}_o$ and $\mathbf{f}_o \sim \int \exp(i\mathbf{k}_o\mathbf{r})\,\mathbf{f}(\mathbf{r})dV$ are the amplitude of the noncollinear mode and the corresponding Fourier-transformed inhomogenity, and the (positive) eigenvalue difference ΔJ describes the relative stability of the noncollinear state.

Equation (7) shows that the noncollinear exchange (ΔJ) suppresses ferromagnetic excitations (a) but enhances and stabilizes the noncollinear mode. In a very large positive field, both $\langle \mathbf{m} \rangle$ and $\mathbf{m}_o$ approach zero, that is, M_z approaches saturation. With reduced field strength h, both volume-averaged random forces, $\langle \mathbf{f} \rangle$, and random-force projections onto the chiral mode, $\mathbf{f}_o$, tend to destabilize the ferromagnetic state. However, due to the opposite sign of the ΔJ contribution, the incommensurate mode is more enhanced, and when $h=DJ/2$, then the saturated or aligned state becomes unstable, and the magnet's spin state switches to a true nonlinear noncollinear state.

IV. DISCUSSION AND CONCLUSIONS

One reason for the complicated spin structure is the nondiagonal character of Q in Eq. (6a). In terms of Fig. 2, spin colloids exhibit a mixing of the m_x and m_y components. The details of the modulation of the noncollinear spin structure are complicated and depend on factors such as the involved materials parameters and the size and geometry of the grains. A simpler but explicated treated case is antiferromagnets with random anisotropy, where $k_o=\pi/a$ and $\mathbf{D}_{ij}=0$.[19] Note that the m_x-m_y mixing is not visible in Eq. (7), due to the projection onto two k-space modes.

In summary, we have investigated how competing interactions affect the spin structure of half-metallic ferromagnets. Our generalized micromagnetic approach reveals that competing exchange interactions yield difficult-to-suppress wave-vector-dependent modulations of the spin structure. In addition, there are small random anisotropy and Dzyaloshinskii–Moriya noncollinearities at the grain and phase boundaries. These noncollinearities lead to spin-colloidal behavior and open a harmful second spin channel in half-metallic ferromagnets.

ACKNOWLEDGMENTS

This work is supported by NSF-MRSEC, NRI, and CMRA.

[1] V. Yu. Irkhin and M. I. Katsnel'son, Phys. Usp. **37**, 659 (1994).
[2] P. A. Dowben and R. Skomski, J. Appl. Phys. **95**, 7453 (2004).
[3] Ch. Hordequin, E. Lelievre-Berna, and J. Pierre, Physica B **234**, 602 (1997).
[4] C. N. Borca *et al.*, Phys. Rev. B **64**, 052409 (2001).
[5] R. Skomski and P. A. Dowben, Europhys. Lett. **58**, 544 (2002).
[6] *Spin Electronics*, edited by M. Ziese and M. J. Thornton (Springer, Berlin, 2001).
[7] P. A. Dowben and R. Skomski, J. Appl. Phys. **93**, 7948 (2003).
[8] R. Skomski and J. M. D. Coey, *Permanent Magnetism* (IOP, Bristol, 1999).
[9] R. Skomski, J. Magn. Magn. Mater. **272–276**, 1476 (2004).
[10] W. Jones and N. H. March, *Theoretical Solid State Physics I* (Wiley, London, 1973).
[11] A. Kashyap, R. Skomski, R. F. Sabiryanov, S. S. Jaswal, and D. J. Sellmyer, IEEE Trans. Magn. **39**, 2908 (2003).
[12] K. Moorjani and J. M. D. Coey, *Magnetic Glasses* (Elsevier, Amsterdam, 1984).
[13] K.-H. Fischer and A. J. Hertz, *Spin Glasses* (Cambridge University Press, Cambridge, 1991).
[14] R. Skomski, J. Phys.: Condens. Matter **15**, R841 (2003).
[15] L. M. Sandratskii, Phys. Rev. B **64**, 134402 (2001).
[16] A detailed proof requires careful summation over all pairs of neighbors while taking into account that $\mathbf{D}_{ij}=-\mathbf{D}_{ji}$.
[17] A. Michels, J. Weissmüller, U. Erb, and J. G. Barker, Phys. Status Solidi A **189**, 509 (2002).
[18] W. F. Brown, *Micromagnetics* (Wiley, New York, 1963).
[19] J. Richter and R. Skomski, Phys. Status Solidi B **153** 711 (1989).