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The origin and physical nature anisotropic exchange interactions is investigated. Emphasis is on nonrelativistic exchange anisotropies, as encountered, for example, in intermetallics with layered crystal structures. The summation of site-resolved exchange interactions is analyzed, and it is shown that Ruderman–Kittel-type long-range exchange yield converging exchange-stiffness expressions down to atomic length scales. In general, the resulting exchange stiffness is anisotropic, even if the interaction is mediated by an isotropic free electron gas. The determination of the mean-field Curie temperature from pair-exchange interactions requires the diagonalization of an interaction matrix, as opposed to simple site averaging. © *2005 American Institute of Physics.* [DOI: 10.1063/1.1850401]

I. INTRODUCTION

Many scientifically interesting and technologically important materials are anisotropic. This includes alloys and oxides with noncubic crystal structure, disordered and partially ordered magnets, magnetic nanostructures, and multilayers. There are several types of exchange anisotropy. Heisenberg exchange has the familiar structure

$$J(\mathbf{R}_i - \mathbf{R}_j)\mathbf{S}_i \cdot \mathbf{S}_j = J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j, \tag{1}$$

where the J_{ij} are site-resolved pair-exchange parameters and $\mathbf{S}_i$ is the spin of the ith atom. The exchange of Eq. (1) is magnetically *isotropic*; that is, coherent rotation of a magnet's spin system does not change the Heisenberg exchange energy. There is, however, a generally very strong bond anisotropy associated with the vectors $\mathbf{R}_{ij} = \mathbf{R}_j - \mathbf{R}_i$.[1] For example, in layered structures, such as YCo_5 and $L1_0$ magnets, intra- and interlayer exchange may be different,[2,3] but the exchange does not depend on whether the magnetization is in-plane or normal to the layers. These interactions are also the main source of spin noncollinearities encountered in elemental rare earths and in magnetoresistive materials, such as NiMnSb.[4]

Past research has lead to a basic understanding of correlation effects and of the relationship between itinerant and localized features of ferromagnets.[5] Recently, it has become possible to determine exchange interactions from first principles,[2,6,7] and to calculate materials properties such as Curie temperature T_C (Ref. 8) and the exchange stiffness[9] for materials of practical interest.

The bond anisotropy (exchange anisotropy) must not be confused with the relatively weak relativistic anisotropies, which involve spin-orbit coupling and depend on the angle between the magnetization and the crystal axes. Examples are the exchange interactions assumed in the Ising and *XY* models, the magnetocrystalline anisotropy, and the unidirectional Dzyaloshinskii–Moriya exchange. Compared to

Heisenberg exchange, relativistic contributions are smaller by a factor of order α^2, where $\alpha = 1/137$ is Sommerfeld's fine structure constant.[10]

The main focus of this paper is to relate site-resolved exchange coefficients J_{ij} to experimental quantities, such as Curie temperature and exchange stiffness. This helps, for example, to identify specific structure-related Curie-temperature contributions. Site-resolved exchange interactions are also important for the understanding of the finite-temperature anisotropy of permanent magnets.[11] The summation over all neighbors i and j is nontrivial,[12,13] particularly in nanostructures,[14] where first-principle calculations are not feasible in the near future.

II. ORIGIN OF EXCHANGE ANISOTROPY

Heisenberg exchange is *isotropic* and, due to its electrostatic nature, relatively strong ($J/k_B \sim 100$ K). A specific example of Heisenberg exchange is the Ruderman–Kittel–Kasuya–Yosida or RKKY exchange between two localized moments. In the simplest case, the theory assumes free electrons, but there are also effective-mass approximations, and asymptotic RKKY-type oscillations are encountered in nanostructures.[13,14] For a free-electron gas of wave vector k_F,

$$J_{ij} = J(|\mathbf{r}_i - \mathbf{r}_j|) = J(R) \sim \cos(2k_F R)/R^3. \tag{2}$$

Due to the isotropy of the underlying free-electron gas, the RKKY interaction is isotropic, $J(\mathbf{r}_i - \mathbf{r}_j) = J(|\mathbf{r}_i - \mathbf{r}_j|)$. However, there is an anisotropic net exchange if the lattice formed by the embedded magnetic moments has a low symmetry.

The relativistic *anisotropic exchange* means that the interaction strength depends on the spin direction relative to the bond vector $\mathbf{R}_i - \mathbf{R}_j$. In principal-axis representations, it can be written as $J_{xx,ij}S_{x,i}S_{x,j} + J_{yy,ij}S_{y,i}S_{y,j} + J_{zz,ij}S_{z,i}S_{z,j}$. The exchange anisotropy is a relatively small relativistic effect; that is, $|J_{x,ij} - J_{z,ij}| \ll J_{x,ij}$ and $|J_{y,ij} - J_{z,ij}| \ll J_{z,ij}$. Since this exchange anisotropy is a small correction to isotropic exchange, it is frequently neglected.

Lowest-order *magnetocrystalline anisotropy*, in principal-axis representation $K_{xx}M_x^2 + K_{yy}M_y^2 + K_{zz}M_z^2$, is rela-

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tivistic as well but it cannot be considered as a small correction to a leading nonrelativistic term. As a consequence, it must be taken into account when the length scale approaches or exceeds $a_o/\alpha=7.2$ nm,[10] for example, when considering magnetic domains.

A third class of relativistic exchange interactions is the *Dzyaloshinskii–Moriya* (or DM) interaction $H_{DM}=-\frac{1}{2}\Sigma_{ij}\mathbf{D}_{ij}\cdot\mathbf{S}_i\times\mathbf{S}_j$, where the vector $\mathbf{D}_{ij}=-\mathbf{D}_{ji}$ reflects the local environment of the magnetic atom.[15] Net DM interactions require local environments with sufficiently low symmetry and occur, for example, in some crystalline materials, such as α-Fe_2O_3 (hematite), in amorphous magnets, spin glasses, and magnetic nanostructures.[10,15,16]

III. EXCHANGE STIFFNESS

On a continuum level, the Heisenberg exchange of cubic materials is described by the energy

$$E_{ex} = \int A[\nabla(\mathbf{M}/M_s)]^2 dV. \tag{3}$$

The *exchange stiffness A* is important for the description of various nanoscale and macroscopic phenomena, such as coercivity and spin waves.[10,11] Due to its continuum character, it may break down on an atomic scale, but even on a nearest-neighbor scale the relative errors are often smaller than 20%.[10,17]

To derive the exchange stiffness from the exchange parameters J_{ij}, we rewrite Eq. (3) in terms of the magnetization angles. Without loss of generality, we keep $\phi=0$, so that

$$E_{ex} = \int A(\nabla\theta)^2 dV. \tag{4}$$

Next, we take into account that

$$E_{ex} \sim \Sigma_{ij}J_{ij}\cos(\theta_i-\theta_j) \approx \Sigma_{ij}J_{ij}[1-(\theta_i-\theta_j)^2/2]. \tag{5}$$

Using the expansion $\theta_j=\theta_i+\nabla\theta\cdot(\mathbf{r}_j-\mathbf{r}_i)$ and comparing the result with Eq. (4) yields $A\sim\Sigma_{ij}J_{ij}(\mathbf{r}_i-\mathbf{r}_j)^2$. This well-known expression has been used to derive A for nearest-neighbor interactions, but it diverges for long-range interactions J_{ij}. An example is the RKKY interaction [Eq. (2)], where integration over all neighbors yields $A\sim\int 1/R^3R^2R^2 dR=\infty$. The reason is that $\theta_j=\theta_i+\nabla\theta\cdot(\mathbf{r}_j-\mathbf{r}_i)$ breaks down for large distances $R=|\mathbf{r}_i-\mathbf{r}_j|$.

To solve the problem, we use the Fourier transform $J_\mathbf{k}$ of $J(|\mathbf{r}-\mathbf{r}'|)$. Since $\int A(\nabla\theta)^2 dV=\int J_\mathbf{k}\theta_\mathbf{k}^2 d\mathbf{k}$ and $\int A(\nabla\theta)^2 dV=-\int Ak^2\theta_\mathbf{k}^2 d\mathbf{k}$, A is given by the quadratic coefficient of the expansion of $J_\mathbf{k}$ with respect to $\mathbf{k}$. With $\mathbf{k}=k\mathbf{e}_k$, $\mathbf{R}=R\cos\theta'\mathbf{e}_k+R\sin\theta'\mathbf{e}_\perp$, and $dV=4\pi R^2\sin\theta' d\theta' dR$, we obtain

$$J_\mathbf{k} \sim \int J(R)\frac{\sin(kR)}{kR}R^2 dR. \tag{6}$$

For RKKY interaction, $J_\mathbf{k}=F(k)$ is the Lindhard screening function.[5,18] In 1952, this function was introduced to describe electron-density oscillations (Friedel oscillations), but it also applies to RKKY oscillations.[5,18] Figure 1 compares the Lindhard function (solid line) with the exchange-stiffness approximation (dashed line). We see that the exchange-
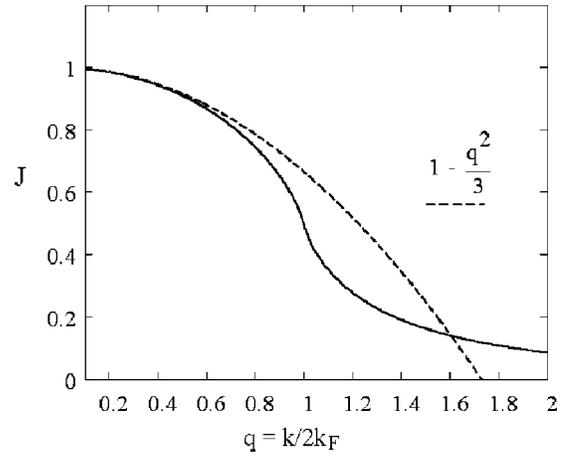

FIG. 1. Exchange energy as a function of the wave vector of the magnetization inhomogenity: Lindhard function (solid line) and exchange-stiffness approximation (dashed line).

stiffness approximation works well for long wavelengths, but breaks down when k becomes comparable to k_F.

In *noncubic* materials, A must be replaced by the 3×3 exchange-stiffness tensor $A_{\mu\nu}$, and the energy is $\Sigma_{\mu\nu}\int A_{\mu\nu}\partial\mathbf{M}/\partial x_\mu\cdot\partial\mathbf{M}/\partial x_\nu dV$. Here, the indices μ and ν (from the middle of the Greek alphabet) denote the spatial coordinates x, y, and z of the bonds. The energy is anisotropic with respect to the nabla operator $\nabla_\mu=\partial/\partial_\mu$ (bond anisotropy), but isotropic with respect to the magnetization $\mathbf{M}$. By contrast, the relativistic anisotropic exchange $\Sigma_{\alpha\beta}\int A_{\alpha\beta}\nabla M_\alpha\nabla M_\beta dV$ is isotropic with respect to ∇, but anisotropic with respect to $\mathbf{M}$.

IV. CURIE TEMPERATURE

For isotropic lattices with nearest-neighbor interactions, the spin-1/2 mean-field Curie temperature is $T_C=zJ/3k_B$, where z is the number of nearest neighbors. A frequently used expression is $T_C=\langle J_o\rangle/3k_B$, where J_o is the single-site exchange and the average is over all lattice sites.[2,12] However, this approach fails when there is a pronounced dispersion of the site-specific $J_o(\mathbf{r}_i)$. An extreme example is a mixture of two ferromagnetic phases with equal volume fractions but different Curie temperatures T_1 and $T_2>T_1$. In the approximation just described, $T_C=(T_1+T_2)/2$, but in reality $T_C=T_2$.[19] The effect persists down to very small length scales and occurs, in a slightly weakened form, even on an atomic scale.[19,20]

The most general mean-field treatment of the critical behavior of ferromagnets is based on the diagonalization of the interaction matrix J_{ij}. The Curie temperature is given by the largest eigenvalues of J_{ij},[19,20] and the corresponding eigenmodes are generally nonuniform. Figure 2 shows a simple two-dimensional example. Site averaging would yield $T_C=(7T_{black}+9T_{white})/16$, but the exact mean-field Curie temperature is obtained by diagonalizing a 6×6 matrix. For $T_{black}=2T_o$ and $T_{white}=T_o/2$, the correct result is $T_C=1.421T_o$, as compared to the site-averaged result $T_C=1.156T_o$.
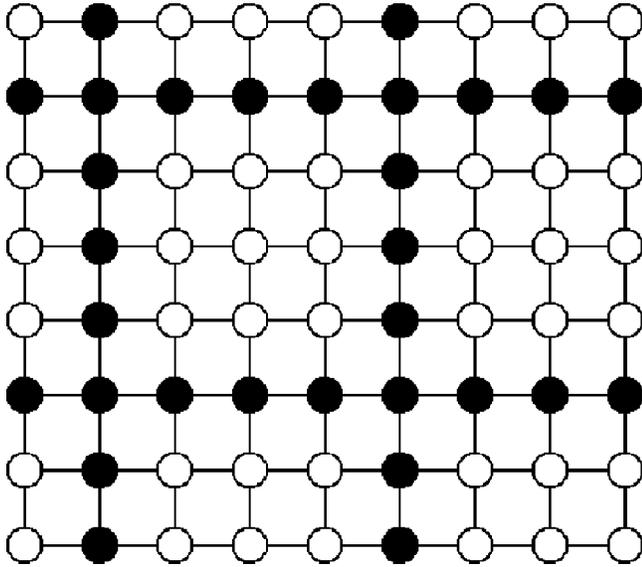
FIG. 2. Mean-field treatment of a simple two-dimensional lattice. There are two types of atoms (black and white) with altogether six nonequivalent sites. T_C is essentially determined by the black atoms. Note that many magnetic compounds can be considered as anisotropic structures where strongly magnetic layers are separated by essentially nonmagnetic layers.

V. DISCUSSION AND CONCLUSIONS

Heisenberg interactions [Eq. (1)] require well-defined atomic magnetic moments, where $\mathbf{S}^2 = S_o^2$. In insulators, $S_o^2 = S(S+1)$, whereas in metals, S_o is an expectation value and $\mathbf{S}/S_o$ has the character of a unit vector that describes the local magnetization direction. In some materials, this is a very crude approximation. Examples are semimetals, such as Sb, exchange-enhanced Pauli paramagnets, such as Pt, and very weak itinerant ferromagnets, such as $ZrZn_2$.[11,21] A similar situation is encountered in $L1_0$ magnets, where the $4d$ or $5d$ moments (Pd or Pt) are induced by the $3d$ atoms (Fe or Co). There the moment of the $4d/5d$ layers exhibits an explicit dependence on the relative spin arrangement of the adjacent $3d$ layers: it is nonzero for ferromagnetic $3d$-$3d$ coupling but zero for antiferromagnetic (AFM) coupling.[3]

A closely related issue is that first-principles calculations based on perturbation theory (force theorem) are non-self-consistent. If an atomic moment $\mathbf{S}_i$ experiences a negative (or AFM) net interaction, then the Heisenberg-type reversal of that atomic moment does not necessarily yield the correct energy, because any significant changes in $\mathbf{S}_i$ leads to a rearrangement of the one-electron levels. These correction are not considered in this paper.

In conclusion, we have investigated the origin and manifestation of anisotropic magnetic interactions, with particular focus on nonrelativistic phenomena. Free-electron RKKY interactions are inherently isotropic, but embedding a low-symmetry lattice of local magnetic moments yields spatially anisotropic exchange interactions. Compared to short-range exchange interactions, the summation over RKKY-type interactions requires specific care, but yields convergent expressions for the exchange stiffness. To determine the mean-field Curie temperature, it is necessary to diagonalize a matrix whose size is given by the number of nonequivalent lattice sites. This Curie temperature is typically larger than that obtained from the volume-averaged exchange.

[1] L. J. de Jongh and A. R. Miedema, Adv. Phys. **23**, 1 (1975).
[2] A. Kashyap, R. Skomski, R. F. Sabiryanov, S. S. Jaswal, and D. J. Sellmyer, IEEE Trans. Magn. **39**, 2908 (2003).
[3] R. Skomski, A. Kashyap, and D. J. Sellmyer, IEEE Trans. Magn. **39**, 2917 (2003).
[4] R. Skomski and P. A. Dowben, Europhys. Lett. **58**, 544 (2002).
[5] P. Fulde, *Electron Correlations in Molecules and Solids* (Springer, Berlin, 1991).
[6] R. F. Sabiryanov and S. S. Jaswal, J. Magn. Magn. Mater. **177–181**, 989 (1998).
[7] V. A. Gubanov, A. I. Liechtenstein, and A. V. Postnikov, *Magnetism and the Electronic Structure of Crystals* (Springer, Berlin, 1992).
[8] H. Zeng, R. Sabirianov, O. Mryasov, M. L. Yan, K. Cho, and D. J. Sellmyer, Phys. Rev. B **66**, 184425 (2002).
[9] K. D. Belashchenko, J. Magn. Magn. Mater. **270**, 415 (2004).
[10] R. Skomski, J. Phys.: Condens. Matter **15**, R841 (2003).
[11] R. Skomski and J. M. D. Coey, *Permanent Magnetism* (Institute of Physics, Bristol, 1999).
[12] V. P. Antropov, B. N. Harmon, and A. N. Smirnov, J. Magn. Magn. Mater. **200**, 148 (1999).
[13] R. Skomski, R. F. Sabiryanov, and S. S. Jaswal, J. Appl. Phys. **85**, 5890 (2000).
[14] R. Skomski, Europhys. Lett. **48**, 455 (1999).
[15] K.-H. Fischer and A. J. Hertz, *Spin Glasses* (Cambridge University Press, Cambridge, UK, 1991).
[16] K. Moorjani and J. M. D. Coey, *Magnetic Glasses* (Elsevier, Amsterdam, 1984).
[17] H. R. Hilzinger and H. Kronmüller, Phys. Lett. **51**A, 59 (1975).
[18] N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Saunders, Philadelphia, 1976).
[19] R. Skomski and D. J. Sellmyer, J. Appl. Phys. **87**, 4756 (2000).
[20] J. S. Smart, *Effective Field Theories of Magnetism* (Saunders, Philadelphia, 1966).
[21] P. Mohn, *Magnetism in the Solid State* (Springer, Berlin, 2003).