Effective spin Hamiltonian for the CuO planes in La_2CuO_4 and metamagnetism

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We show that the effective spin Hamiltonian used previously to describe the CuO planes of La_2CuO_4 does not lead to a net ferromagnetic moment for CuO planes and hence does not describe the metamagnetic behavior seen experimentally. We construct for the first time a Hamiltonian from the symmetries of the crystal structure which does lead to metamagnetism. The linear spin-wave spectrum is also calculated. This work points to the necessity of constructing effective spin Hamiltonians for metamagnetic systems which have the same symmetries as the system they are to describe.

We construct for the first time an effective spin Hamiltonian for the CuO planes of undoped La_2CuO_4 whose classical ground state has a small ferromagnetic moment and calculate the corresponding linear spin-wave spectrum. The CuO planes are known to have a small ferromagnetic moment from the metamagnetic behavior seen in measurements of the static magnetic susceptibility,[1] although the interactions are predominantly antiferromagnetic. This weak ferromagnetism (WF) is present in other materials and Dzyaloshinskii[2] proposed that this could be accounted for by the presence of an extra term in the Hamiltonian beyond the isotropic antiferromagnetic (AF) Heisenberg term of the form $\mathbf{D}\cdot\mathbf{S}_i\times\mathbf{S}_j$, where $\mathbf{S}_i$ and $\mathbf{S}_j$ are spins at the sites i and j. He pointed out that this contribution is not forbidden by symmetry in an expansion of the free energy if the symmetry of the system is sufficiently low. Moriya[3] then showed that this extra term arises from the effect of the spin-orbit interaction on the superexchange characterized by the Heisenberg J. He showed that $|\mathbf{D}|\sim(\Delta g/g)J$, where g is the value of the free-electron gyromagnetic ratio and Δg was the shift in that value due to the spin-orbit interaction. He also gave rules for determining the direction of $\mathbf{D}$ from the symmetries of the spin system.

This extra term, the Dzyaloshinskii-Moriya (DM) term, has been applied by a number of authors[1-10] to the description of WF and has more recently been used to describe the metamagnetism[1] or spin-flop transition,[7] the magnetoresistance,[7] and the conductivity[8] of undoped La_2CuO_4. In the present work we show that the inclusion of the original DM term, where $\mathbf{D}$ is taken to be a constant, in an effective spin Hamiltonian does not lead to a description of WF but that a generalization of the DM term, which is determined by the symmetry properties of the crystal structure, does lead to a net ferromagnetic moment in the ground state. We consider as an explicit example the CuO planes of La_2CuO_4 and show that the important symmetry of the crystal structure which leads to the CuO planes having a net ferromagnetic moment is that each Cu site should be a center of inversion. We note in passing that an alternative model for WF was introduced by Borovik-Romanov and Orlova[11] and is discussed in detail in Ref. 4. In this model WF arises because there are different g tensors for different sublattices. The WF is only manifest in applied fields and since there is evidence for WF in the absence of applied magnetic fields in the systems being considered here we shall not discuss this model further.

Since the discovery of high-temperature superconductivity in a number of different families of compounds, all of which have in common CuO planes, there has been a great deal of work done to characterize and understand the properties of the CuO planes in these compounds. The simplest of these compounds are derived from La_2CuO_4 by doping either with strontium[12] or with barium[13] or excess oxygen.[14] The unit cells of these La_2CuO_4 compounds contain a single CuO plane and the essential physical properties which lead to high-temperature superconductivity should be found in this system. An important part of the investigation of these compounds is the study of undoped La_2CuO_4. Undoped La_2CuO_4 undergoes a structural phase transition from tetragonal to orthorhombic symmetry at a temperature around 500 K. At the transition temperature the CuO octahedra tilt by about 0.027 rad so that the CuO planes are buckled.[15,16] This is shown in Fig. 1, which is taken from Endoh *et al.*[17] This system and the closely related compound, K_2NiF_4, have been studied by x-ray diffraction and neutron scattering and recently reviews of theoretical and experimental work on this system by Chakravarty[18] and Birgeneau and Shirane[19] have appeared.

In order to find a spin Hamiltonian to describe the CuO planes of La_2CuO_4 one has to determine the symmetries of the crystal structure. In particular, one can arrange the tilted CuO octahedra, shown in Fig. 1, in two different periodic arrays as shown in Fig. 2. We will consider the arrangement of Fig. 2(a) here although there is some evidence of a low-temperature tetragonal phase[20] which may be represented by the arrangement of Fig. 2(b). The arrangement in Fig. 2(a) can be arrived at by projecting the CuO octahedra, as shown in Fig. 2 of Grande *et al.*,[16] onto the CuO planes and all our analysis is based on this arrangement of Cu and O atoms. We will describe this system by a spin Hamiltonian which is made

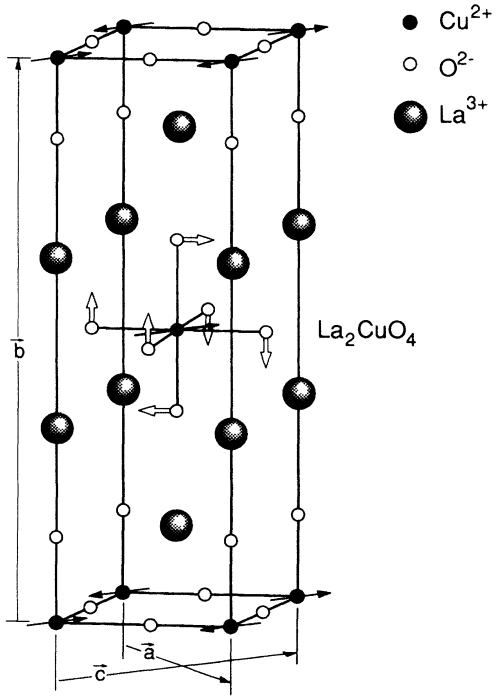

FIG. 1. Crystal structure of undoped La_2CuO_4 [taken from Endoh *et al.* (Ref. 14)] where the arrows indicate the tilting of the CuO octahedra.

up of an isotropic AF Heisenberg term together with a term of the Dzyaloshinskii-Moriya (DM) type which is shown in Eq. (1).

$$H = \sum_{\langle i,j \rangle} \{J\mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j\} , \tag{1}$$

where i and j are nearest-neighbor sites.

This is a more general form of DM term than has been used by previous authors[2-10] in that $\mathbf{D}_{ij}$ varies from bond to bond, and we will show below that this form is essential in order to describe either La_2CuO_4 or weak ferromagnetism in general. The effective spin Hamiltonian must have the same symmetries as the system and in particular the DM term must be invariant under transformations which leave the crystal structure unchanged. Considering Fig. 2(a) we find the following symmetry operations. (i) Twofold axis of rotation normal to the plane through a point C midway between each two Cu spins, (ii) a center of inversion at each Cu spin, i.e., the point B, (iii) reflection in a diagonal through the plane containing AD and the normal to the CuO plane, (iv) twofold rotational axis through BF, and (v) a center of inversion through the center of each plaquette O. The symmetry (i) implies, following the rules originally found by Moriya, that the DM vector $\mathbf{D}_{ij}$ lies in the CuO plane. This may be seen by considering the contribution to the Hamiltonian from the interaction of the spins at A and F and remembering that under the symmetry (i) the two components of $\mathbf{S}$ in the plane change sign whereas the com-

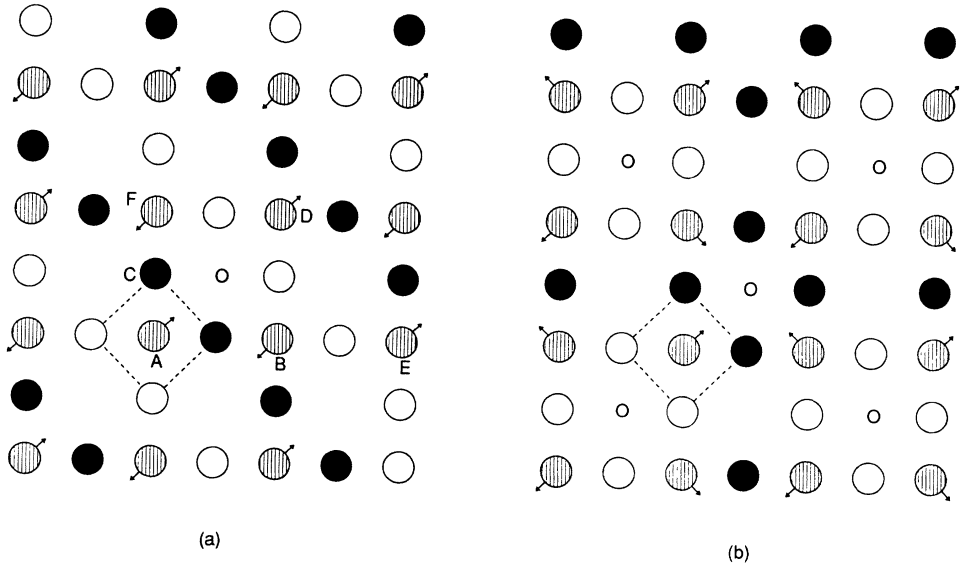

FIG. 2. (a) The "orthorhombic" arrangement of the CuO octahedra. Here a schematic drawing of a CuO plane in the orthorhombic arrangement is shown which may be found by considering Fig. 2 of Grande *et al.* (Ref. 14) and projecting the tilted octahedra on to the CuO plane. The dashed line connects the face centered O atoms associated with the octahedron shown in Fig. 1. The shaded circles are O atoms which are tilted down out of the plane, while the open circles are O atoms tilted up out of the plane. The hatched circles are the sites of the Cu atoms and of the spins. They are also the sites of the O atoms in the CuO plane above that under consideration, and the arrows indicate the direction of motion for the O atoms associated with the tilt of the octahedra. In this arrangement diagonal lines of octahedra tilt in the same direction. (b) The "tetragonal" arrangement of the octahedra. The symbols are the same as in (a). In this arrangement the lattice may be broken into sets of four octahedra which successively tilt towards their common axis (the line through the point O) or away from it.

ponent parallel to the axis of rotation remains unchanged. The symmetry (ii) implies that the DM vector alternates in sign on successive bonds. This may be seen by considering the contribution to the Hamiltonian from the DM interaction between the spins at A, B, and E,

$$\mathbf{D}_{AB}\cdot\mathbf{S}_A\times\mathbf{S}_B+\mathbf{D}_{BE}\cdot\mathbf{S}_B\times\mathbf{S}_E \ . \tag{2}$$

Under inversion A and E are swapped and B does not change. $\mathbf{S}_A$, $\mathbf{S}_B$, and $\mathbf{S}_E$ also remain unchanged under inversion so that Eq. (2) becomes

$$\mathbf{D}_{AB}\cdot\mathbf{S}_E\times\mathbf{S}_B+\mathbf{D}_{BE}\cdot\mathbf{S}_B\times\mathbf{S}_A \ . \tag{3}$$

In order for Eqs. (2) and (3) to be identical, as required by the inversion symmetry of the crystal structure $\mathbf{D}_{AB}=-\mathbf{D}_{BE}$. Applying (iii) one finds that if $\mathbf{D}_{AB}=(d_1,d_2,0)$, then $\mathbf{D}_{AF}=(-d_2,-d_1,0)$, and applying (iv) one finds that $\mathbf{D}_{AB}=-\mathbf{D}_{FD}$ and $\mathbf{D}_{AF}=-\mathbf{D}_{BD}$. Not all of the above symmetries are independent. For instance (iii) after (v) is the same as (iv) and there are other symmetries such as mapping rows onto one another which are made up of (i) to (v). The constraints that symmetries of the kind (ii)–(v) place on $\mathbf{D}_{ij}$ have not previously been discussed in the literature to our knowledge. Once $\mathbf{D}_{ij}$ is fixed on a single bond, these symmetries determine $\mathbf{D}_{ij}$ on the entire lattice.

d_1 and d_2 remain to be determined from a microscopic calculation; however, two cases may be identified, (1) $\mathrm{sgn}(d_1)\neq\mathrm{sgn}(d_2)$ and (2) $\mathrm{sgn}(d_1)=\mathrm{sgn}(d_2)$. The ground state for case (1) has a net ferromagnetic moment, whereas that for case (2) does not. If J in Eq. (1) is set to zero, case (1) has a ground-state spin configuration with two sublattices, and case (2) has four sublattices. The ground-state spin configuration does not change as d_1/d_2 is varied, except that there is a first-order (discontinuous) transition when the case switches between (1) and (2) (either d_1 or d_2 is equal to zero). There is always a Goldstone mode associated with a global rotation of the spin configuration about a particular axis. That axis is $(1,-1,0)$ for case (1) and (1,1,0) for case (2). The ground state is in general frustrated, in that it is impossible to construct a state for the extended system with the energy per bond as low as $-|\mathbf{D}_{ij}|$. The only exceptions are when $d_1=-d_2$ and $d_1=d_2$, which are unfrustrated. Since case (2) does not describe systems with a nonzero ferromagnetic moment, we consider hereafter only case (1). We consider the representative example $d_1=-d_2$, leaving a more detailed investigation of case (1) to be reported elsewhere.[21]

Aside from the global rotational invariance about the DM vector, $(1,-1,0)$, the (unfrustrated) ground state is uniquely determined, and is shown in Fig. 3. The spins lie in the plane perpendicular to the DM vector which we henceforth refer to as the DM plane. There is a net ferromagnetic moment due to the DM vector. The existence of the net moment relies on the fact that the DM vector alternates in sign from one bond to the next. This property of the DM vector is missing in the work of previous authors[1–10] where $\mathbf{D}$ was taken to be a constant. The classical ground state for that case is shown in Fig. 4 and one sees that the direction of the spin spirals so that

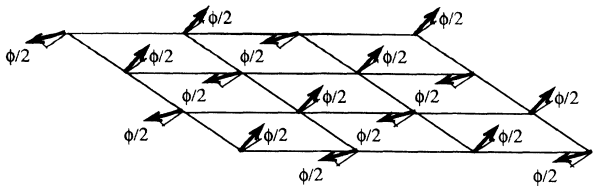

FIG. 3. The classical ground state for the Hamiltonian where the DM vector alternates in sign on neighboring bonds. All spins lie in the DM plane.

there is no net moment. Previous authors concluded that there was a net moment by assuming that there were only two sublattices and then analyzing the free energy of the system in terms of the magnetizations of these sublattices. Clearly there are more than two sublattices, and, in fact, unless

$$\phi=\tan^{-1}(|\mathbf{D}_{ij}|/J)=\pi/n \ ,$$

where n is an integer, there are an infinite number of sublattices. So taking $\mathbf{D}_{ij}$ to be a constant does not lead to a ferromagnetic moment and such a Hamiltonian cannot describe WF or metamagnetic systems. This point was hinted at by Keffer[22] in a review of spin waves some time ago, however the implications for descriptions of WF have been ignored up to now so that recently, experimental results[7–10] have been analyzed keeping $\mathbf{D}_{ij}$ constant.

Given the classical ground state it is straightforward to calculate the spin-wave spectrum in the mean-field approximation. The ground state may be described in terms of two sublattices, A and B, and the directions of the sublattice magnetizations specify directions of spin, $\mathbf{S}_A^0$ and $\mathbf{S}_B^0$. Deviations of $\mathbf{S}_A$ and $\mathbf{S}_B$ from these directions correspond to excitations of the system. The equations of motion for $\mathbf{S}_A$ and $\mathbf{S}_B$ are linearized with respect to these deviations and one finds for the spin-wave spectrum

$$\omega(\mathbf{q})=4S\sqrt{[J_{\mathrm{DM}}+Jf(\mathbf{q})]\{J_{\mathrm{DM}}[1-f(\mathbf{q})]\}} \ , \tag{4}$$

where

$$J_{\mathrm{DM}}=\sqrt{J^2+|\mathbf{D}_{ij}|^2}$$

and

$$f(\mathbf{q})=\tfrac{1}{2}[\cos(q_x a)+\cos(q_y a)] \ .$$

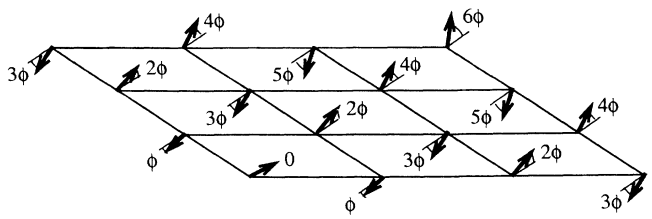

FIG. 4. The classical ground state for the Hamiltonian where the DM vector is constant for each bond. All spins lie in the DM plane.

In Fig. 5 the spectrum is plotted for different values of the ratio $|\mathbf{D}_{ij}|/J$ and one sees that the effect of the DM term is to open a gap at $\mathbf{q}=(\pi/a,\pi/a)$ because it now costs energy for the spins to have a component out of the DM plane. One finds that in the long-wavelength limit $\omega(\mathbf{q})=|\mathbf{q}|V_{sp}$ with a renormalized spin-wave velocity V_{sp}. The spin-wave spectrum for the case of constant $\mathbf{D}$ can be calculated despite the fact that there may be an infinite number of sublattices. An orthonormal triad of vectors $(\mathbf{S}_j^0,\mathbf{S}_j^1,\mathbf{S}_j^2)$ are defined at each site j, with $\mathbf{S}_j^0$ in the direction of the spin in the classical ground state, S_j^1 in the direction of $\mathbf{D}_{ij}$, and $\mathbf{S}_j^2=\mathbf{S}_j^0\times\mathbf{S}_j^1$. In this basis, the equations of motion at each site are identical. One could also have derived $\omega(\mathbf{q})$ for the alternating $\mathbf{D}_{ij}$ case, Eq. (4), using this procedure. One finds that $\omega(\mathbf{q})$ is given by Eq. (4) in the constant $\mathbf{D}$ case also. The spin-wave spectra are the same for both constant and for alternating $\mathbf{D}_{ij}$ because the DM vector enters quadratically in the calculation.

In the above analysis it was found that the ferromagnetic moment of the CuO planes could point in any direction within the DM plane which is the origin of the Goldstone mode leading to $\omega(\mathbf{q})$ vanishing in the long-wavelength limit. However, it is known from neutron-scattering experiments that the spins have a definite direction. This may be incorporated into the spin Hamiltonian by making the Heisenberg part of the Hamiltonian anisotropic in the DM plane thus breaking the rotational symmetry about the DM vector of the ground-state spin configuration. There is a weak effective antiferromagnetic coupling between the CuO planes, J_p, which leads to the ferromagnetic moments of adjacent CuO planes being antiferromagnetically aligned. (The interplanar coupling J_p is added phenomenologically for a cubic lattice. The real situation is more complicated, since the Cu sites in adjacent planes are not directly above one another.) These extra terms modify the Hamiltonian for the system so that it becomes

$$H=\sum_{\langle i,j\rangle}\{J_1[S_i^xS_j^x+S_i^yS_j^y]+J_2S_i^zS_j^z + d_{ij}[(S_i^x+S_i^y)S_j^z-S_i^z(S_j^x+S_j^y)]\} + \sum_{\langle k,k'\rangle}J_p\mathbf{S}_k\cdot\mathbf{S}_{k'}\,, \tag{5}$$

where $\mathbf{D}_{ij}=d_{ij}(1,-1,0)$, i and j are nearest-neighbor sites in the same CuO plane and k and k' are nearest-neighbor sites in adjacent CuO planes. The metamagnetic behavior then follows when a sufficiently strong magnetic field is applied that the planar moments all align rather than alternate. This metamagnetism depends on $\mathbf{D}_{ij}$ changing sign on alternating bonds, which gives the ferromagnetic moment to begin with, and on the anisotropy of the Heisenberg term in the DM plane, which picks out a preferred direction for the moments.

Following the same procedure as for the two-dimensional case we calculate the spin-wave spectrum and find

$$\omega(Q)=\{[\Omega_{1\rho}(q)+\Omega_{1z}(q_z)][\Omega_{2\rho}(q)+\Omega_{2z}(q_z)]\}^{1/2} \tag{6a}$$

where

$$\Omega_{1\rho}(\mathbf{q})=4S\left[\left[J'_{\rm DM}+\frac{J_1+J_2}{2}f(\mathbf{q})\right] + \frac{(J_1-J_2)}{2}\frac{(J_1+J_2)}{2J'_{\rm DM}}[1+f(\mathbf{q})]\right]\,, \tag{6b}$$

$$\Omega_{1z}(q_z)=2SJ_p[1+g(q_z)]\,, \tag{6c}$$

$$\Omega_{2\rho}(\mathbf{q})=4S\left[\frac{J_1-J_2}{2}[1+f(\mathbf{q})]+J'_{\rm DM}[1-f(\mathbf{q})]\right]\,, \tag{6d}$$

$$\Omega_{2z}(q_z)=2SJ_p[1-g(q_z)]\,, \tag{6e}$$

$$g(q_z)=\cos(q_zb)\,, \tag{6f}$$

and the effective coupling constant in the DM plane now becomes

$$J'_{\rm DM}=\left[\left[\frac{(J_1+J_2)^2}{4}\right]+|\mathbf{D}_{ij}|^2\right]^{1/2}\,. \tag{6g}$$

Equations (6) are written for the extended zone but one may easily recover the corresponding expressions for the reduced zone in which there are two branches of the spin-wave spectrum by folding back the spectrum into the reduced zone. Mermin and Wagner[23] showed that if there was a continuous symmetry in the ground state of a

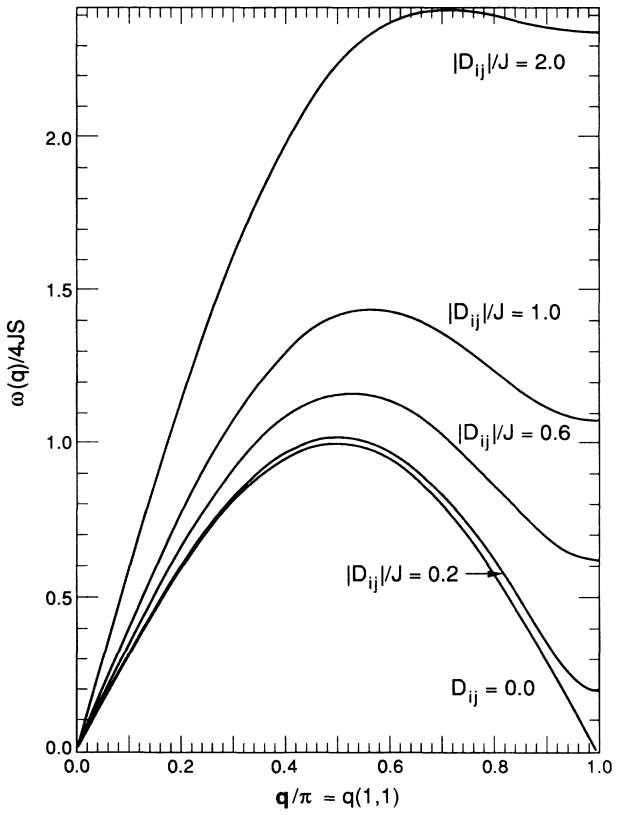

FIG. 5. Spin-wave spectrum along the (1,1) direction, Eq. (4).

two-dimensional system, thermal fluctuations destroy the long-range order. In the case of the Hamiltonian in Eq. (1) describing uncoupled planes there is a Goldstone mode in the DM plane so that there is no long-range order at finite temperatures. However, if a magnetic field is applied which has a component in the DM plane a direction is picked out for the ferromagnetic moment and the continuous symmetry no longer exists. As a result a gap opens up at $\mathbf{q}=\mathbf{0}$ in the spin-wave spectrum and there is long-range order up to a finite temperature. A similar situation arises for the $J_1 \neq J_2$ in Eq. (5) even if $J_p=0$ and there is no applied magnetic field. Examining Eqs. (6) one sees that the anisotropy in the DM plane leads to a gap in the spectrum at $\mathbf{q}=\mathbf{0}$ whether or not the planes are coupled. In particular, the expressions for the two gaps which appear in the reduced zone are found by evaluating the $\omega(\mathbf{Q})$ at (0,0,0) and $(\pi,\pi,0)$. One finds

$$\omega(0,0,0)=4S\left\{(J_1-J_2)\left[\frac{J_1+J_2}{2}\left[1+\frac{(J_1-J_2)}{J'_{\mathrm{DM}}}\right]+J'_{\mathrm{DM}}+J_p\right]\right\}^{1/2}, \tag{7a}$$

$$\omega(\pi,\pi,0)=4S\{J'_{\mathrm{DM}}[2J'_{\mathrm{DM}}-(J_1+J_2)+J_p]\}^{1/2}, \tag{7b}$$

which agrees with the results of previous authors,[5,9,22] once J_p is set to zero, $|\mathbf{D}_{ij}|$ is taken to be negligible compared to J_1 or J_2 and $J_1 \approx J_2$. Consequently, the effects of fluctuations, both quantum and thermal, are reduced even in the absence of coupling between planes and, in particular, there is long-range order for sufficiently low temperatures even for the purely two-dimensional case. We are presently carrying out estimates of the effects of fluctuations and the results will be reported elsewhere.

In summary, we have shown that one has to generalize the term originally introduced by Dzyaloshinskii and Moriya in order to describe WF in predominantly antiferromagnetic systems. This WF is a prerequisite, together with an anisotropy of the Heisenberg term in the DM plane, for the metamagnetic behavior seen in La_2CuO_4. In particular, we have shown that when one considers the symmetries of the crystal structure of La_2CuO_4 the DM vector alternates in sign from one bond to the next and it is this property which leads to weak ferromagnetism in the system but which is missing from all previous Hamiltonians used to describe WF. In general, in order to describe the metamagnetic behavior, which is seen in heavy-fermion and other systems,[24] it is necessary to determine the DM vector from the crystal symmetries.

Note added in proof. We are grateful to P. Kumar for bringing to our attention the work of Q. Xia and P. S. Riseborough, J. Appl. Phys. **67**, 5478 (1990), on the spin-wave excitation spectrum of a one-dimensional Dzyaloshinski-Moriya antiferromagnet in which the DM alternated in sign from bond to bond.

ACKNOWLEDGMENTS

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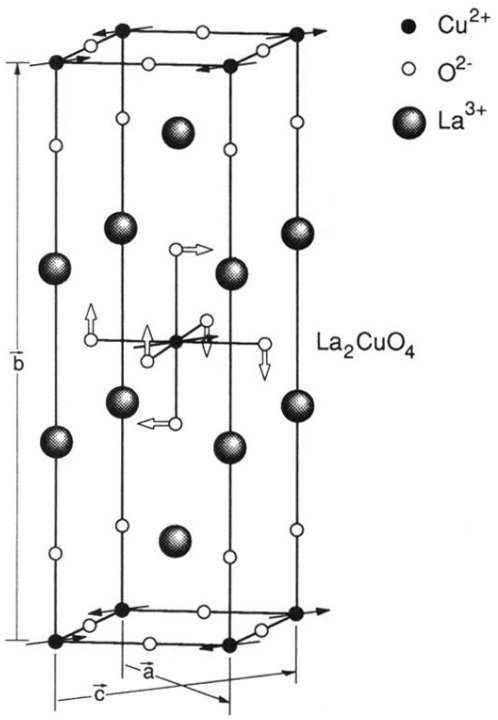

FIG. 1. Crystal structure of undoped La_2CuO_4 [taken from Endoh *et al.* (Ref. 14)] where the arrows indicate the tilting of the CuO octahedra.