## Moriya's Anisotropic Superexchange Interaction, Frustration, and Dzyaloshinsky's Weak Ferromagnetism

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Moriya's expression for the single-bond anisotropic superexchange interaction is shown to possess an overlooked hidden symmetry, isomorphic to the symmetry of the isotropic case. For the unfrustrated case, this symmetry results in a degeneracy of the macroscopic state, implying no unique value for the Dzyaloshinsky weak ferromagnetic moment. A unique value emerges from superexchange *only* when more than a single bond is considered and *only* as a result of frustration. This implies that the symmetric part of the superexchange anisotropy tensor must vary among the bonds. The results are particularly relevant for the spin anisotropies in  $La_2CuO_4$ .

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More than thirty years ago Dzyaloshinsky [1] pointed out that weak ferromagnetism of various mainly antiferromagnetic compounds can be explained by an antisymmetric spin-spin interaction. He showed that the term  $\mathbf{D}^{D}$  ( $\mathbf{M}_1 \times \mathbf{M}_2$ ), allowed in the thermodynamic potential of a sufficiently low-symmetry crystal, favors a canted spin arrangement over the antiferromagnetic one. Here  $\mathbf{D}^{D}$  is the constant Dzyaloshinsky vector and  $\mathbf{M}_{1}$  and  $\mathbf{M}_{2}$ denote the sublattice magnetizations. The microscopic basis for the Dzyaloshinsky conjecture was given by Moriya's [2,3] extension of the Anderson theory [4,5] of superexchange (or "kinetic exchange" [6]) to include spin-orbit coupling. Moriya calculated the tensor describing anisotropic superexchange of two neighboring spins  $S(\mathbf{R})$  and  $S(\mathbf{R}')$  and showed that it contains an antisymmetric part  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$   $\mathbf{S}(\mathbf{R}) \times \mathbf{S}(\mathbf{R}')$ . Moriya suggested that this term represents the leading anisotropy, because it is linear in the spin-orbit coupling while the symmetric anisotropies are of second-order in that coupling. The antisymmetric exchange has recently been observed in the high-temperature superconducting parent material  $La_{2}CuO_{4}$  [7].

The aim of this Letter is to point out that the symmetric anisotropies cannot be neglected in comparison with the antisymmetric ones. We show that the complete expression [2,3] for the two-spin anisotropic superexchange can be mapped via a gauge transformation, onto the isotropic Anderson [4] Hamiltonian. As a result, states with different ferromagnetic moments are shown to be degenerate with the purely antiferromagnetic state. Thus the anisotropy of the two-spin superexchange interaction does not lift the degeneracy of the corresponding ground state. Considering only the superexchange interaction, the degeneracy is lifted only as a result of frustration of this gauge transformation over the entire lattice. This happens when the vectors  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$  (Moriya vectors below) vary from bond to bond in a nontrivial way. Thus, frustration is a necessary condition for the anisotropic superexchange interaction to explain an observable weak ferromagnetism with some definite net ferromagnetic moment. We also show that one of the principal axes of the symmetric part of Moriya's one-bond superexchange anisotropy tensor is always parallel to the corresponding Moriya vector. This yields the surprising result that symmetric superexchange anisotropies vary from bond to bond with the variation of the Moriya vectors. Finally, we show how frustration yields weak ferromagnetism in  $La_2CuO_4$ .

Moriya derived his expression by extending Anderson's formalism [4] of superexchange interaction. He started from the one-electron Hamiltonian

$$H = \sum_{\mathbf{R}} \sum_{\sigma} \varepsilon(\mathbf{R}) a_{\sigma}^{\dagger}(\mathbf{R}) a_{\sigma}(\mathbf{R}) + \sum_{\mathbf{R} \neq \mathbf{R}'} \sum_{\sigma} b(\mathbf{R} - \mathbf{R}') a_{\sigma}^{\dagger}(\mathbf{R}) a_{\sigma}(\mathbf{R}') + \sum_{\mathbf{R} \neq \mathbf{R}'} \sum_{\sigma\sigma'} a_{\sigma}^{\dagger}(\mathbf{R}) [\mathbf{C}(\mathbf{R} - \mathbf{R}') \cdot \boldsymbol{\sigma}]_{\sigma\sigma'} a_{\sigma'}(\mathbf{R}') , \qquad (1)$$

where b and C are the transfer integrals [3],  $a^{\dagger}$  and a are the electron creation and annihilation operators, and  $\sigma$  is the vector of Pauli spin matrices. The first two terms in Eq. (1) represent Anderson's one-electron Hamiltonian [4]. The third term, which is nondiagonal in spin space, takes account of the spin-orbit interaction. Following Anderson, Moriya used second-order perturbation calculations to derive his expression for the interaction between the spins at **R** and **R'** [Eqs. (2.3) and (2.4) in Ref. [3]],

$$E_{\mathbf{R},\mathbf{R}'}^{(2)} = J_{\mathbf{R},\mathbf{R}'} \mathbf{S}(\mathbf{R}) \cdot \mathbf{S}(\mathbf{R}') + \mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M} \cdot \mathbf{S}(\mathbf{R}) \times \mathbf{S}(\mathbf{R}') + \mathbf{S}(\mathbf{R}) \vec{\Gamma}_{\mathbf{R},\mathbf{R}'} \mathbf{S}(\mathbf{R}') .$$
(2)

This expression was derived [2,3] under the assumption that the ground state of each ion is nondegenerate except for being a Kramer's doublet, and we will confine our analysis below just to this spin- $\frac{1}{2}$  case. The coefficients in (2) are given by [8]

$$J_{\mathbf{R},\mathbf{R}'} = (4/U) |b(\mathbf{R} - \mathbf{R}')|^2, \qquad (3a)$$

$$\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M} = \frac{4i}{U} [b(\mathbf{R}-\mathbf{R}')\mathbf{C}(\mathbf{R}'-\mathbf{R}) - b(\mathbf{R}'-\mathbf{R})\mathbf{C}(\mathbf{R}-\mathbf{R}')],$$
(3b)

$$\vec{\Gamma}_{\mathbf{R},\mathbf{R}'} = \frac{4}{U} [\mathbf{C}(\mathbf{R}-\mathbf{R}')\mathbf{C}(\mathbf{R}'-\mathbf{R}) + \mathbf{C}(\mathbf{R}'-\mathbf{R})\mathbf{C}(\mathbf{R}-\mathbf{R}')]$$

$$-\widetilde{IC}(\mathbf{R}-\mathbf{R}')\cdot\mathbf{C}(\mathbf{R}'-\mathbf{R})], \qquad (3c)$$

where U is the energy required to put two electrons on the

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same ion (the Hubbard energy). Equation (3b) defines the Moriya vector. Equation (3c) gives the anisotropy tensor  $\vec{\Gamma}$ ,  $\vec{I}$  being the unit matrix. Note that Eq. (3a) contains the numerical factor 4, which differs from the factor 2 in Moriya's Eq. (2.4a) [3].

Moriya argued that the order of magnitude of the vector  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$  is  $(\Delta g/g)J_{\mathbf{R},\mathbf{R}'}$ , and that of the symmetric tensor  $\overline{\Gamma}_{\mathbf{R},\mathbf{R}'}$  is  $(\Delta g/g)^2 J_{\mathbf{R},\mathbf{R}'}$ , where g is the gyromagnetic ratio and  $\Delta g$  its deviation from the free electron value. Therefore  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$  seems to be the leading-order anisotropy. It is customary to assume (see, for example, Ref. [9]) that this implies that in the classical ground-state spin configuration the spins are confined to the plane perpendicular to  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$ . The role of the symmetric anisotropy  $\vec{\Gamma}_{\mathbf{R},\mathbf{R}'}$ is to choose a particular direction in that plane. We would like to stress that although  $\overline{\Gamma}_{\mathbf{R},\mathbf{R}'}$ , is indeed small, it still plays as important a role as does  $D_{\mathbf{R},\mathbf{R}'}^{M}$ . The reason is that in discussing the anisotropies one has to compare not  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$  and  $\vec{\Gamma}_{\mathbf{R},\mathbf{R}'}$  but rather  $(\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M})^2$  and  $J_{\mathbf{R},\mathbf{R}'}$ .  $\vec{\Gamma}_{\mathbf{R},\mathbf{R}'}$ . Obviously, the latter are of the same order of magnitude. Much more surprising is the fact that for the superexchange interaction the values of the two corresponding anisotropy parameters are exactly equal to one another. To see this we note that for a specific bond C(R - R') $= [C(R'-R)]^*$  and  $b(R-R') = [b(R'-R)]^*$ , and in the case of nondegenerate ground-state orbitals the transfer integral b is real while C is purely imaginary [3]. It is now convenient to define

 $C(R - R') = i\hat{d}(R - R')|C(R - R')| = -C(R' - R), \quad (4)$ 

so that  $\hat{\mathbf{d}}(\mathbf{R} - \mathbf{R}')$  is a unit vector along the  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$  direction [see Eq. (3b)]. With these notations the energy (2) takes the form

$$E_{\mathbf{R},\mathbf{R}'}^{(2)} = \frac{4}{U} \left( b^2 \mathbf{S}(\mathbf{R}) \cdot \mathbf{S}(\mathbf{R}') + 2bC \hat{\mathbf{d}} \cdot \mathbf{S}(\mathbf{R}) \times \mathbf{S}(\mathbf{R}') + C^2 \left\{ 2[\hat{\mathbf{d}} \cdot \mathbf{S}(\mathbf{R})][\hat{\mathbf{d}} \cdot \mathbf{S}(\mathbf{R}')] - \mathbf{S}(\mathbf{R}) \cdot \mathbf{S}(\mathbf{R}') \right\} \right),$$
(5)

where for clarity we have put  $\hat{\mathbf{d}}(\mathbf{R}-\mathbf{R}') \equiv \hat{\mathbf{d}}$ ,  $b(\mathbf{R}-\mathbf{R}') \equiv b$ , and  $|\mathbf{C}(\mathbf{R}-\mathbf{R}')| \equiv C$ . It follows from Eq. (5) that one of the principal axes of the tensor  $\vec{\Gamma}_{\mathbf{R},\mathbf{R}'}$  is directed along the  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$ . Indeed [see Eq. (5)], the spin components along the  $\hat{\mathbf{d}}$  direction do not couple to those perpendicular to  $\hat{\mathbf{d}}$ . It also follows that the corresponding principal value is the largest, and that the two other values are equal to one another and therefore cannot account for any anisotropy in the plane perpendicular to  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$ . Moreover, expression (5) is just the *scalar* product of two spins,  $\mathbf{S}'(\mathbf{R})$  and  $\mathbf{S}'(\mathbf{R}')$  obtained from the original ones by rotations around the  $\hat{\mathbf{d}}$  axis with angles  $-\theta$  and  $\theta$ , respectively, where  $\tan\theta = C/b$ . Hence the energy is

$$E_{\mathbf{R},\mathbf{R}'}^{(2)} = \left(J_{\mathbf{R},\mathbf{R}'} + \frac{|\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{\mathcal{M}}|^2}{4J_{\mathbf{R},\mathbf{R}'}}\right) \mathbf{S}'(\mathbf{R}) \cdot \mathbf{S}'(\mathbf{R}') , \qquad (6)$$

where

$$S'(\mathbf{R}) = (1 - \cos\theta) [\mathbf{d} \cdot S(\mathbf{R})] \hat{\mathbf{d}} + \cos\theta S(\mathbf{R}) - \sin\theta S(\mathbf{R}) \times \hat{\mathbf{d}},$$
(7)  
$$S'(\mathbf{R}') = (1 - \cos\theta) [\hat{\mathbf{d}} \cdot S(\mathbf{R}')] \hat{\mathbf{d}} + \cos\theta S(\mathbf{R}')$$

Since (6) is invariant under rotations of all the  $S'(\mathbf{R})$ 's, it follows that also the Hamiltonian (2) does not confine the original spins  $S(\mathbf{R})$  to any particular direction and therefore it does not choose any particular ferromagnetic moment in the classical ground state. One can find in this ground state configurations with a net magnetic moment ranging continuously from zero (spins parallel to  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$ ) up to  $\pm \sin\theta$  (spins perpendicular to  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$ ). Note that we have an unusual situation. The interaction is anisotropic—the vector  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$  does represent a special direction in the space of the original spins, identifying the special case with no net ferromagnetic moment. This vector also determines the local rotation, and therefore the symmetry of the states in the original spin variables. However, this anisotropy does not lift the degeneracy of the ground state-there is no energy of anisotropy. Clearly, in order to map Eq. (2) onto Eq. (6) it is necessary to take into account the tensor  $\overrightarrow{\Gamma}_{\mathbf{R},\mathbf{R}'}$ , even though it is second order in  $\Delta g/g$  and, last, but not least, to replace the factor 2 in Moriya's expression for  $J_{\mathbf{R},\mathbf{R}'}$  by the correct factor 4.

In the traditional approach, one first ignores the last term in Eq. (2), and notes that  $D_{R,R'}^{M}$  plays two roles: It generates a ferromagnetic moment perpendicular to  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$ and to the local staggered moment L = S(R) - S(R'), and it contributes an anisotropy energy of order  $|\mathbf{D}_{\mathbf{R},\mathbf{R}'}^M|^2/J$  which prefers L to be perpendicular to  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^M$ . One then switches on the symmetric anisotropy and that chooses alternative orientations for L. The ferromagnetic moment vanishes when the symmetric anisotropy prefers  $L \parallel D_{\mathbf{R},\mathbf{R}'}^{\mathbf{M}}$ . Our calculation, which includes only the superexchange effects, shows that the last term in (2) exactly cancels the above-mentioned anisotropy energy, so that all the directions of L (and therefore also many values of the ferromagnetic moment) have the same energy. Indeed, any additional anisotropy may pick a direction for L. As we show below, frustration picks this direction even without additional anisotropies.

Quite naturally, the question arises as to whether the disappearance of the anisotropic terms is accidental and occurs only in second-order perturbation theory. To explore this possibility, we return to the one-bond part of the one-electron Hamiltonian (1). For convenience, we rewrite it in the form

$$H_{\mathbf{R},\mathbf{R}'} = \varepsilon(\mathbf{R}) \sum_{\sigma} a_{\sigma}^{\dagger}(\mathbf{R}) a_{\sigma}(\mathbf{R}) + \varepsilon(\mathbf{R}') \sum_{\sigma} a_{\sigma}^{\dagger}(\mathbf{R}') a_{\sigma}(\mathbf{R}') + \sum_{\sigma\sigma'} a_{\sigma}^{\dagger}(\mathbf{R}) (te^{i\theta\hat{\mathbf{d}}\cdot\boldsymbol{\sigma}})_{\sigma\sigma'} a_{\sigma'}(\mathbf{R}') + \text{H.c.}, \qquad (8)$$

where t,  $\theta$ , and  $\hat{\mathbf{d}}$  depend upon  $\mathbf{R} - \mathbf{R}'$ ,  $t \cos\theta = b$ , and  $t \sin\theta = C$ . From the form (8) it is clear that by performing a unitary transformation of the operators,

$$a_{\sigma}(\mathbf{R}) \rightarrow \hat{\mathbf{a}}_{\sigma}(\mathbf{R}) = \sum_{\sigma'} [e^{-i(\theta/2)\hat{\mathbf{d}} \cdot \sigma}]_{\sigma\sigma'} a_{\sigma'}(\mathbf{R}) ,$$
  
$$a_{\sigma}(\mathbf{R}') \rightarrow \hat{\mathbf{a}}_{\sigma}(\mathbf{R}') = \sum_{\sigma'} [e^{i(\theta/2)\hat{\mathbf{d}} \cdot \sigma}]_{\sigma\sigma'} a_{\sigma'}(\mathbf{R}') ,$$
  
(9)

the one-bond Hamiltonian (8) is transformed into

$$H_{\mathbf{R},\mathbf{R}'} = \varepsilon(\mathbf{R}) \sum_{\sigma} \hat{\mathbf{a}}_{\sigma}^{\dagger}(\mathbf{R}) \hat{\mathbf{a}}_{\sigma}(\mathbf{R}) + \varepsilon(\mathbf{R}') \sum_{\sigma} \hat{\mathbf{a}}_{\sigma}^{\dagger}(\mathbf{R}') \hat{\mathbf{a}}_{\sigma}(\mathbf{R}') + \sum_{\sigma} t [\hat{\mathbf{a}}_{\sigma}^{\dagger}(\mathbf{R}) \hat{\mathbf{a}}_{\sigma}(\mathbf{R}') + \hat{\mathbf{a}}_{\sigma}^{\dagger}(\mathbf{R}') \hat{\mathbf{a}}_{\sigma}(\mathbf{R})].$$
(10)

This is exactly the one-bond part of the Anderson Hamiltonian [4], from which one derives the *isotropic* superexchange. Note that the spin-orbit coupling appears here as a modification on the transfer integral: Instead of *b* (the transfer integral in the absence of spin-orbit coupling) there is now  $t=b/\cos\theta$  ( $\theta=0$  in the absence of spin-orbit coupling). Note also the one-to-one correspondence between the transformations described by Eqs. (9) and (7).

Thus we have shown that for one bond, the above mapping is correct to all orders in perturbation theory. We stress that the possibility to transform Eq. (8) into Eq. (10) does not depend on the explicit expressions in Eq. (3) and, hence, confirms our claim about the factor 4 in Eq. (3a). Note also that even in the case of chargetransfer insulators, when one has to explicitly eliminate electron degrees of freedom on the ligand ions, the effective Hamiltonian for hopping between magnetic ions has the form (8), allowing for the above mapping.

We turn next to the superexchange spin Hamiltonian for the entire lattice. Let us point out that, while the direction of the Dzyaloshinsky vector  $\mathbf{D}^{D}$  is determined by the symmetry of the entire system [1], the direction of the Moriya vector  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$  is determined by the transfer integrals  $b(\mathbf{R} - \mathbf{R}')$  and  $C(\mathbf{R} - \mathbf{R}')$  which to leading order do not depend on the symmetry of the lattice as a whole (but they do depend, of course, on the symmetry of the particular bond [3]). The directions of the Moriya vectors for different bonds on the lattice are related to each other by the lattice symmetry [9,10]. A priori, these vectors need not all be along the same direction, nor along special lattice directions. The principal axes of the symmetric parts of the various one-bond anisotropy tensors also need not be the same for all the bonds. Note that the direction and the absolute value of the Moriya vector for a specific bond determine the direction and the absolute value of the rotation necessary to transform the corresponding one-bond part of the spin Hamiltonian to the isotropic form [see Eqs. (7) and (9)]. Clearly, if the product of four such rotations around each square plaquette is equal to unity, then it is possible to transform the total superexchange spin Hamiltonian to the isotropic form, Eq. (6). The degeneracy of the corresponding ground state is lifted only when the total transformation is *frustrated* (the product of four rotations around the plaquette is not equal to unity).

Consider first the unfrustrated case, in which the rotations of spins at all the sites are compatible with each other. In this case, the complete Hamiltonian maps onto a sum over the isotropic interactions of Eq. (6). For each bond in the classical ground state  $S'(\mathbf{R})$  and  $S'(\mathbf{R}')$  are antiparallel and there is an infinite degeneracy for all the possible directions of S'. However, not all directions are equivalent. If the spins  $S'(\mathbf{R}) = -S'(\mathbf{R}')$  are along the Moriya vector  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$  [which is parallel to  $\hat{\mathbf{d}}$  of Eq. (7)], then it follows from Eq. (7) that also  $S(\mathbf{R}) = -S(\mathbf{R}')$ , and hence there is no net ferromagnetic moment. For all other spin directions (which have the same energy),  $S(\mathbf{R})$ is not antiparallel to  $S(\mathbf{R}')$ , and a net ferromagnetic moment follows. However, this moment depends on the angle between the spins and  $D^{M}_{R,R'}$  [the terms  $S \times \hat{d}$  in Eq. (7)] and is in practice determined by an anisotropy arising from other sources (e.g., direct exchange, dipolar interactions, an external magnetic field, etc.). In fact, the degeneracy is expected to yield a very strong sensitivity to external fields, with diverging susceptibilities. It would be very interesting to find materials which have such an unfrustrated structure.

In the *frustrated* case, there exists no rotation of the spins that will map the complete Hamiltonian onto an isotropic one. Thus, frustration lifts the degeneracy of the ground state. It breaks the symmetry and picks a specific configuration, with a definite net ferromagnetic moment, by minimizing the complete Hamiltonian.

For the two-sublattice case, the Moriya Hamiltonian (2) is easily mapped onto the Dzyaloshinsky thermodynamic potential

$$\mathbf{\Omega} = J\mathbf{M}_1 \cdot \mathbf{M}_2 + \mathbf{D}^D \cdot \mathbf{M}_1 \times \mathbf{M}_2 + \mathbf{M}_1 \vec{\Gamma} \mathbf{M}_2.$$
(11)

Here,

$$J \equiv J_{\mathbf{R},\mathbf{R}'}, \quad \vec{\Gamma} \equiv \frac{1}{N_0} \sum_{l} \vec{\Gamma}_{\mathbf{R},\mathbf{R}'}$$
(12a)

are independent of the direction of the bonds (i.e., the sign of  $\mathbf{R} - \mathbf{R}'$ ), while [11]

$$\mathbf{D}^{D} \equiv \frac{1}{N_0} \sum_{\mathbf{R}'} \mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}, \qquad (12b)$$

where **R** and **R'** belong to sublattices 1 and 2, respectively, and  $N_0$  is the coordination number. It follows that the degeneracy under rotations of **M**<sub>1</sub> and **M**<sub>2</sub> persists on the macroscopic level only in the special case when all the  $\mathbf{D}_{\mathbf{K},\mathbf{R}'}^{L}$  (for fixed **R**) are equal to each other, so that  $\mathbf{D}^{D} \equiv \mathbf{D}_{\mathbf{K},\mathbf{R}'}^{R}$ . We next show that this is equivalent to the absence of frustration. Consider the plaquette with the sites  $1 \rightarrow 2 \rightarrow 3 \rightarrow 4 \rightarrow 1$ . The degeneracy implies that  $[9] \mathbf{D}_{12}^{H} = \mathbf{D}_{14}^{H} = -\mathbf{D}_{41}^{H}$  and  $\mathbf{D}_{34}^{H} = \mathbf{D}_{32}^{H} = -\mathbf{D}_{23}^{H}$ . Therefore,  $\mathbf{D}_{12}^{H} + \mathbf{D}_{23}^{H} + \mathbf{D}_{34}^{H} = 0$ . Thus, the product of the rotations in Eq. (9) around the plaquette is equal to unity and there is no frustration.

Hence, one can identify  $\mathbf{D}_{\mathbf{K},\mathbf{R}'}^{M}$  and  $\mathbf{D}^{D}$  only when all the Moriya vectors have the same magnitude and alternate in sign on successive bonds along each path on the

lattice ["canonical Dzyaloshinsky-Moriya (DM) antiferromagnet"]. As we already noted, the corresponding classical ground-state manifold cannot be characterized by any definite net ferromagnetic moment, because these moments differ for different states belonging to the manifold. To break this hidden symmetry and to pick up some configuration with a definite moment it is necessary to introduce frustration. This happens only if one deviates from the "canonical" case, so that the Moriya vectors are not identical to the Dzyaloshinsky vector. Hence, a *nontrivial difference* between the Dzyaloshinsky and the Moriya vectors is a necessary condition for the anisotropic superexchange interaction to explain an observable weak ferromagnetism with some specific value of the net ferromagnetic moment.

However, this condition is not sufficient. A simple counterexample arises when the Moriya vectors are all equal to each other along the path encircling the basic plaquette. In this case, Eq. (12b) yields  $D^{D}=0$ , while  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M} \neq 0$ . A ferromagnetic moment arises only if  $\mathbf{D}^{D} \neq 0$ . Let us illustrate this point on a specific example [10] related to the magnetic properties of the CuO<sub>2</sub> planes in the orthorhombic phase of La<sub>2</sub>CuO<sub>4</sub>. In this case, the symmetry of each bond is sufficiently low [3] to allow  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M} \neq 0$ . At the same time, the symmetry of the crystal structure is sufficiently high to imply that all the Moriya vectors are of the same magnitude. Thus, it is sufficient in this case to consider the frustration resulting from the relative orientations of various  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$ 's, avoiding the complications connected with their magnitude. Each Cu-Cu bond in the  $CuO_2$  plane possesses a twofold symmetry axis perpendicular to the plane. Since the corresponding Moriya vector must be perpendicular to this axis [3], it follows that all the  $D_{\mathbf{R},\mathbf{R}'}^{M}$ 's are confined to the plane. The general orientation of the Moriya vectors is a superposition of two unique cases in which all the  $D_{R,R'}^{M}$ 's are aligned along the same direction [9,10]: In case 1, the  $\mathbf{D}_{\mathbf{R},\mathbf{R}'}^{M}$ 's are aligned along the orthorhombic  $\hat{\mathbf{a}}$  axis, and in case 2, they are aligned along the orthorhombic  $\hat{c}$  axis (in the notation of Ref. [12]). In case 1 [9,10], all the Moriya vectors alternate in sign on successive bonds along any path. Thus it represents a "canonical" DM antiferromagnet. In contrast, in case 2 the Moriya vectors, though alternating in sign along straight paths, have the same sign along the path encircling the basic plaquette [10,13]. In this case, Eq. (12b) yields  $\mathbf{D}^{D} \equiv 0$  and the spins order antiferromagnetically without any ferromagnetic moment. Weak ferromagnetism originates only when (i) the projections of the Moriya vectors on the  $\hat{a}$ axis have the same (nonzero) magnitude and alternate their sign from bond to bond, and (ii) these vectors also have nonzero components along the  $\hat{c}$  axis, which do not alternate in sign along the path encircling the basic plaquette. In a separate paper [10] we show that the onebond superexchange Moriya vectors are almost perpendicular to the corresponding bonds, i.e., are directed neither along the  $\hat{c}$  nor along the  $\hat{a}$  axis. Only because of this fact is it possible to explain [10] the observable weak ferromagnetism of La<sub>2</sub>CuO<sub>4</sub> on the basis of the theory of superexchange interactions. The resulting mean-field spin Hamiltonian is identical to the one used phenomenologically by Thio *et al.* [7] to account for the peculiar magnetic properties of La<sub>2</sub>CuO<sub>4</sub>, assuming that one takes for the value of  $J^{bc}$  of Thio *et al.* the magnitude of the projections of the Moriya vectors onto the  $\hat{a}$  axis.

We emphasize that only the superexchange (or "kinetic exchange" [6]) interaction has been considered. It is believed [3-5] that this interaction dominates exchange in insulators. Other interactions (e.g., direct or "potential" [6] exchange, dipole-dipole interactions) are not as symmetric as the superexchange and provide (along with the frustration of the one-bond superexchange interaction) another channel of lifting the degeneracy of the groundstate spin configurations.

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