Comment on "Moriya's Anisotropic Superexchange Interaction, Frustration, and Dzyaloshinsky's Weak Ferromagnetism"

In a recent Letter [1], Shekhtman, Entin-Wohlman, and Aharony have shown that the single-bond anisotropic superexchange interaction [2] does not provide anisotropy, and that frustration is a necessary condition for yielding weak ferromagnetism in La_2CuO_4. Their arguments have been based entirely on the single-band Hubbard model. In this Comment, we would like to point out that, in a realistic model with multiorbitals on the ligand oxygen ions, the interaction between the two spins on the neighboring Cu ions is already anisotropic, and that this single-bond anisotropy, rather than frustration among the bonds, is in fact the origin of the weak ferromagnetism in La_2CuO_4.

A relevant model, which one starts with, must contain in-plane oxygen orbitals coming into play due to lattice distortions. Let us write the one-hole Hamiltonian

$$\begin{aligned} H=&\sum_{\mathbf{R}'}\sum_{n}\sum_{\sigma}\varepsilon_n(\mathbf{R}')p^{\dagger}_{n\sigma}(\mathbf{R}')p_{n\sigma}(\mathbf{R}') \\ &+\sum_{\mathbf{R}\neq\mathbf{R}'}\sum_{n}\sum_{\sigma}[b_n(\mathbf{R}-\mathbf{R}')d^{\dagger}_{\sigma}(\mathbf{R})p_{n\sigma}(\mathbf{R}')+\text{H.c.}] \\ &+\sum_{\mathbf{R}\neq\mathbf{R}'}\sum_{n}\sum_{\sigma\sigma'}\{d^{\dagger}_{\sigma}(\mathbf{R})[\mathbf{C}_n(\mathbf{R}-\mathbf{R}')\cdot\sigma]_{\sigma\sigma'}p_{n\sigma'}(\mathbf{R}')+\text{H.c.}\} \end{aligned} \tag{1}$$

in an extended notation of Ref. [1]. $\mathbf{R}$ runs over the Cu sites and $\mathbf{R}'$ runs over the O sites. $d^{\dagger}_{\sigma}(\mathbf{R})$ creates a hole with spin σ in the ground-state $3d$ orbital at site $\mathbf{R}$, and $p^{\dagger}_{n\sigma}(\mathbf{R}')$ creates a hole with spin σ in the in-plane oxygen $2p_n$ ($n=x,y,z$) orbital at site $\mathbf{R}'$, where the z axis is taken perpendicular to the CuO_2 plane. It is then proved, by following a recipe of Ref. [1], that the one-bond part of the Hamiltonian (1) is isotropic *only if* $\mathbf{C}_n(\mathbf{R}-\mathbf{R}')/b_n(\mathbf{R}-\mathbf{R}')$ is independent of n, i.e., in this case the unitary transformation of operators

$$\begin{aligned} d_{\sigma}(\mathbf{R}_1)\rightarrow\hat{\mathbf{d}}_{\sigma}(\mathbf{R}_1)&=\sum_{\sigma'}[e^{-i(\theta/2)\hat{\mathbf{c}}\cdot\sigma}]_{\sigma\sigma'}d_{\sigma'}(\mathbf{R}_1)\,, \\ d_{\sigma}(\mathbf{R}_2)\rightarrow\hat{\mathbf{d}}_{\sigma}(\mathbf{R}_2)&=\sum_{\sigma'}[e^{i(\theta/2)\hat{\mathbf{c}}\cdot\sigma}]_{\sigma\sigma'}d_{\sigma'}(\mathbf{R}_2)\,, \end{aligned} \tag{2}$$

transforms the one-bond Hamiltonian into the isotropic one, where $\hat{\mathbf{c}}$ denotes a unit vector in the direction of $\mathbf{C}_n(\mathbf{R}_1-\mathbf{R}')/b_n(\mathbf{R}_1-\mathbf{R}')$, and $\mathbf{R}_1$ and $\mathbf{R}_2$ are the neighboring Cu sites between which the O site $\mathbf{R}'$ locates. In the real situations, however, $\mathbf{C}_n(\mathbf{R}_1-\mathbf{R}')/b_n(\mathbf{R}_1-\mathbf{R}')$ cannot be independent of n. We thus have *an anisotropic single bond.* In Ref. [3], we have derived the full spin Hamiltonian including the oxygen $2p_z$ orbitals and found that, in the low-temperature orthorhombic phase of La_2CuO_4, the single-bond anisotropy is characteristic of the weak-ferromagnetic spin canting.

Shekhtman, Entin-Wohlman, and Aharony [1] argued that their Hamiltonian can be mapped onto the Dzyaloshinsky thermodynamic potential via Eqs. (12a) and (12b) of Ref. [1], and thus the weak-ferromagnetic ordered state is realized in the presence of the frustration. We have reexamined the ground-state spin structure of the system by a finite-size exact diagonalization technique [3]. It is found that for their Hamiltonian (i.e., in the system without the effect of the $2p_z$ orbitals), the frustration works to suppress the correlation for the uniform magnetization rather than to raise it. Thus, the ground state of this two-dimensional quantum system seems not to follow the behavior suggested in Ref. [1]. However, for the Hamiltonian including the $2p_z$-orbital contribution, a strong enhancement of the correlation for the uniform magnetization is observed, which straightforwardly leads to the emergence of the weak ferromagnetism. The single-bond anisotropy due to the oxygen multiorbitals is thus essential. The frustration among different bonds seems insufficient. The weak ferromagnetism recently observed in the low-temperature tetragonal phase of $La_{2-x}Nd_xCuO_4$ [4] can also be explained in terms of the single-bond anisotropy due to the $2p_z$-orbital contribution [3]; it should be noted that the frustration among different bonds has no possibility for yielding the weak ferromagnetism in this phase [5].

Finally, let us note that our arguments are not only relevant for the La_2CuO_4-type crystals, but are to imply generally that when one studies the magnetism which arises from spin-orbit coupling under distorted lattices, it is of essential importance to examine contributions from multiorbitals of the ligand ions.

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