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Yildirim et al. Reply

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Abstract

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Yildirim et al. Reply: The Skanthakumar et al. Comment [1] draws attention to the existing evidence in favor of the noncollinear spin structure of Nd$_2$CuO$_4$ (NCO), and we agree that this evidence is compelling [2]. The aim of our Letter [3] was to understand microscopic origins of interactions that would stabilize the three-dimensional spin structures of various cuprates. As noted in Ref. [23] of [3], the occurrence of domains creates difficulties in separating the collinear and noncollinear structures. Being aware of the experiments cited in Ref. [1], we followed Matsuda et al. [4] and exploited the former possibility. However, most of the arguments presented in [3] remain correct, and apply equally to both structures. Furthermore, although we successfully explained the collinear structure of many cuprates, the structure proposed in [4] for NCO did not fit into our explanation. As we discuss below, an extension of our argument could supply a microscopic explanation for the observed noncollinear structure.

Contrary to the impression given in the Comment, the “models” in Refs. [5–7] are only phenomenological. They use a symmetry analysis to write down the same types of terms as we derived in [3], but they do not discuss the microscopic origin of each term or include information about their signs and magnitudes. These are crucial in determining the ground state structures. Thus, the models could only list all the possible structures, but could not “already correctly give the noncollinear spin structure as the ground state.” A specific example concerns the dipolarlike terms of the form $-A \sin(\theta_m + \theta_n)$ [Eqs. (9) and (11) in [3]]. Both the collinear and noncollinear structures (Fig. 2 in [3] and Fig. 1 in [1]) require that $A < 0$. This sign was not explained by Ref. [5]. In [3] we showed that, in fact, $A = A_0' - D$, where $A_0' > 0$ is due to the dipolar interactions and the new parameter $D$ results from the anisotropy of the interplanar exchange [Eq. (7) in [3]]. For the Cu-Cu exchange in, e.g., Sr$_2$CuCl$_2$O$_2$, $D$ is small and $A > 0$. We expect the same to happen for Pr$_2$CuO$_4$ (PCO), since the ground state of Pr$^{3+}$ is nonmagnetic. Indeed, this is consistent with both the collinear structure of the PCO discussed in [3] and its noncollinear counterpart suggested in [1]. However, the anisotropy of the Cu-Nd exchange may be enhanced due to the magnetic ground state of Nd$^{3+}$, and this may yield a larger $D$ and $A < 0$.

To distinguish between the collinear and the noncollinear structures, we considered in [3] a single-ion term of the form $-K_{\text{in}} \cos(4\theta_m)$ [Eq. (5) in [3]], which arose from a novel mechanism for systems with isotropic spins $\frac{1}{2}$, namely, zero point spin-wave fluctuations. Although this term prefers ordering along [100] and [010], as in the noncollinear structure, it was too small for the non-rare-earth cuprates. Therefore, all of those cuprates order collinearly along the [110] direction [8]. However, the crystal field splitting on the rare-earth ions (Pr or Nd) should introduce a relatively strong single-ion anisotropy. Indeed, measurements of the single-ion susceptibility of the Pr$^{3+}$ ions in PCO show [9] that their spins prefer ordering along [100] or [010]. Calculations of the anisotropy energies (and of $D$) for the Nd system are currently being performed, but it is reasonable to expect this effect to be even larger than for Pr, since the Nd ground state is magnetic. Given that the Nd spins order along [010] or [100], the Cu-Nd exchange would generate a similar effective single-ion anisotropy on the Cu ions. This could enhance our parameter $K_{\text{in}}$ and yield the observed noncollinear structure. The competition between these energies and the Nd-Cu exchange might also supply an explanation for the successive spin-orientation transitions in NCO [1,4]. Direct measurements of the single-ion susceptibility for Nd$^{3+}$, or of the in-plane spin-wave gaps associated with $K_{\text{in}}$ [Eq. (6) in [3]], could confirm these scenarios.

In conclusion, our Letter provides a microscopic understanding of the three-dimensional spin structures of the non-rare-earth tetragonal cuprates. Our theory can also explain the noncollinear structure of systems such as NCO or PCO, provided that one includes the effects of the strong single-ion anisotropy on the rare-earth ion.

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[2] Similar conclusions were reported by D. Petitgrand et al., in International Conference on Magnetism, Warsaw, 1994.