Comment on "Competition between Helimagnetism and Commensurate Quantum Spin Correlations in $LiCu_2O_2$"

In a neutron scattering investigation of $LiCu_2O_2$ Masuda *et al.* [1] reported the direct observation of an incommensurate (IC) magnetic structure below 22 K. Though this study confirms similar indirect IC observations [2–4] pointing to the presence of frustrated magnetic interactions, they deserve now more detailed work to elucidate the microscopic origin of that frustration. We will show that the adopted antiferromagnetic (AFM) double-chain (DC) Heisenberg model [1–3] [Fig. 1(a)] suggests an unrealistic frustration scenario for $LiCu_2O_2$. It should be replaced by a ferromagnetic-(FM)-AFM frustrated *single*-chain model [Fig. 1(b)] [4,5]. Based on electronic structure (LDA) and cluster calculations as well as a phenomenological analysis of the magnetic susceptibility $\chi(T)$, we arrive at opposite estimates compared with Ref. [1] with respect to the magnitude or sign of the main couplings. The controversy concerns the following main points:

(i) Most importantly, the signs of the nearest-neighbor (NN) inchain exchange J_1 are opposite: AFM + 1.68 meV in Ref. [1] versus FM − 11 ± 3 meV in our analysis [4,7]. For CuO_2 chains with Cu-O-Cu bond angles γ near 94° as in Li_2CuO_2 (with FM inchain order), according to the Kanamori-Goodenough rule and to the FM direct Cu $3d$-O $2p$ exchange, a total FM $J_1 < 0$ can be expected. However, its magnitude is sensitive to the competition with a γ-dependent AFM contribution to J_1 [4]. Hence, too simplified distance-only based suggestions [1] that $|J_1| \gg J_2$ do not hold here.

(ii) We found the NN inchain coupling J_2 AFM (generic for CuO_2 chains), i.e., frustrated with FM J_1 and any J_{DC}. Moreover, we estimated $\alpha \equiv J_2/J_1 \sim -1$. However, the real source of frustration J_2 is ignored in Ref. [1]. Also the $\chi(T)$ (Fig. 1) and the AFM Curie-Weiss constant [2] can be explained with $\alpha = -1.0$ and -1.1, respectively.

(iii) A dominant interchain coupling $J_{DC} \approx 5.8$ meV is claimed by Masuda *et al.* whereas from our LDA analysis a tiny $J_{DC} \sim 0.5$ meV only follows. We ignore it to first approximation. The weakness of J_{DC} is caused by the tiny interchain (DC) overlap of the predominant O $2p_{x,y}$ orbitals of the CuO_4 plaquettes forming the CuO_2 chains. Note that if $J_1 < 0$, the DC is *unfrustrated* for $J_2 = 0$.

Finally, we note that Masuda *et al.* [1] argue that their propagation vector ζ would contradict our J ratio: $\alpha = -1/[4\cos(2\pi\zeta)]$. However, this simple expression is valid for single chains with classical spins $s \gg 1$. In our case with $s = 1/2$, quantum fluctuations [6], interchain coupling, and spin anisotropy do affect the helix and α strongly.

To conclude, the application of the AFM DC-model of Ref. [1] to $LiCu_2O_2$ is not justified whereas the proposed frustrated single-chain model with FM J_1 and AFM J_2 couplings is consistent with the experimental data and the general physics of CuO_2 chains.

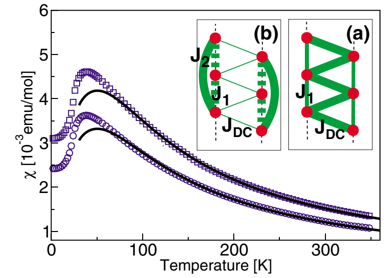

FIG. 1 (color online). Susceptibility of Heisenberg rings with $-J_1 = J_2 = 8.2$ meV, $J_{DC} = 0$. $N = 16$ sites, $g_L = 2.24$ and 2.0, respectively, (full lines) compared with Ref. [1]: □ magnetic field $\mathbf{H} \parallel c$; ∘ $\mathbf{H} \parallel (a, b)$. Inset: the DC scenario [(a), [1]] compared with the single-chain scenario (b). Thick lines symbolize strong coupling. The empirical J's are in accord with LDA and microscopic estimates [7]. Naturally, the finite cluster approach cannot describe the low-T behavior of $\chi(T)$.

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[5] The propagation vector $q = (0.5, \zeta = 0.174, 0)$ measured in standard units $2\pi/b$ of the crystal structure yields a pitch angle $\Phi = \pm 2\pi\zeta + 2\pi n$, $n = 0, 1, 2, \ldots$, within the "FM" interval $-\pi/2 \leq \Phi \leq \pi/2$ [6]. However, within the system of coordinates adopted in Ref. [1] with a lattice constant of $b/2$ there are two solutions $\Phi_{DC} = \pm\pi(n \pm \zeta)$ with "AFM" pitch angles for odd (even) n, respectively, depending on the sign of J_{DC}.
[6] R. Bursill *et al.*, J. Phys. Condens. Matter **7**, 8605 (1995), Fig. 2.
[7] From mapping low-lying magnetic excitations of the five-band Cu $3d$ O $2p$ extended Hubbard model (fitted to spectroscopic data) onto excitations of the Heisenberg model.

