Comment on “Competition between helimagnetism and commensurate quantum spin correlations in LiCu$_2$O$_2$"  

In a neutron scattering investigation of LiCu$_2$O$_2$ Masuda et al. reported the direct observation of an incommensurate (IC) magnetic structure below 22 K. Though this study confirms similar indirect IC observations 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 (Fig. 1a) suggests an unrealistic frustration scenario for LiCu$_2$O$_2$. It should be replaced by a ferromagnetic (fm)-afm frustrated single-chain model (Fig. 1b). Based on electronic structure (LDA) and cluster calculations as well as a phenomenological analysis of magnetic data, we arrive at opposite estimates compared with Masuda et al. with respect to the magnitude/sign of the main couplings. The controversy concerns the following main points:

(i) Most importantly, the signs of the n.n. inchain exchange $J_1$ are opposite: afm $+1.68$ meV in Ref. 1 vs. fm $-11.3$ meV in our analysis. For CuO$_2$ chains with Cu-O-Cu bond angles $\gamma$ near $94^\circ$ as in Li$_2$CuO$_2$ (with fm inchain order), according to the Kanamori-Goedenuogh rule and to the fm direct Cu 3$d$-O 2$p$ exchange, a total fm $J_1 < 0$ can be expected. However, its magnitude is sensitive to the competition with a $\gamma$-dependent afm contribution to $J_1$. Hence, to simplified distance-only based suggestions that $|J_1| >> J_2$, do not hold here.

(ii) We found the nnn inchain coupling $J_2$ afm (generic for CuO$_2$ chains), i.e., frustrated with fm $J_1$ and any $J_{DC}$. Moreover we estimated $J_{2\tau} \sim |J_1|$. However, the important source of frustration $J_2$ is ignored in Ref. 1.

(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. It can be neglected to first approximation. The weak $J_{DC}$ is caused by the tiny interchain (DC) overlap of the predominant O 2$p_{x,y}$ orbital of the CuO$_4$ plaquettes forming the CuO$_2$ chains. Note, that if $J_1 < 0$, the DC is unfrustrated for $J_2=0$.

With $J_1 \sim 11$ meV, we explain also the measured magnetic susceptibility $\chi(T)$ (Fig. 1) and the afm Curie-Weiss constants $\Theta_{CW}$. Approximating the main couplings between two nn-chains $J_1$ in the ab-plane in mean-field theory and the inchain couplings exactly in large clusters, we derive for the bulk value $\Theta_{CW} \approx \Theta_{CW;1D} = -zJ_1/4k_B$, where $z=2$ is the number of interchain n.n.s. From cluster studies we obtained an afm $\Theta_{CW;1D} \approx -42$K for single chains with frustrating afm $J_2 > 0$ and $\alpha = J_2/J_1 = -1.1$. With $J_1 \approx 5.7$ meV one arrives at $\Theta_{CW} = -75$K close to experimental values $\approx -80$ to $-90$K.

Finally, we note that Masuda et al. argue that their propagation vector $\zeta$ 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, interchain coupling, and spin-anisotropy do affect $\alpha$ strongly.

To conclude, the application of the afm DC-model of Ref. 1 to LiCu$_2$O$_2$ 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 generally accepted CuO$_2$-chain physics.

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1 T. Masuda et al., Phys. Rev. Lett. 92, 177201 (2004).
2 S. Zvyagin et al., Phys. Rev. B 66, 064424 (2000).
3 K.-Y. Choi et al., Phys. Rev. B 69, 104421 (2004).
4 A.A. Gippius et al., Phys. Rev. B 70, 024406 (2004).
5 The observed propagation vector $q = (0.5, \zeta = 0.174, 0)$ measured in standard units $2\pi/b$ of the crystal structure yields a pitch angle $\Phi = \pm 2\pi \zeta + 2\pi n, n=0,1,2,...$, in the "fm" interval $-\pi/2 \leq \Phi \leq \pi/2$. However, within the system of coordinates adopted in Ref. 1 with a lattice constant $b/2$ 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 The error bars stems from uncertainties in the screening of bare fm interactions.
7 From mapping low-lying magnetic excitations of the 5-band Cu 3$d$ O 2$p$ extended Hubbard model (fitted to spectroscopic data) onto excitations of the Heisenberg model.
8 Notice the "+"-sign convention of $\Theta_{CW}$ used in Ref. 2.
9 R. Bursill et al., J. Phys. C, 7, 8605 (1995), Fig. 2.
10 Strong interchain exchange may even prevent a helix for-

![FIG. 1: Susceptibility of Heisenberg rings with $-J_1 = J_2 = 8.2$ meV, $J_{DC}=0$. $N=16$ sites, and Lande-Factors $g_\parallel = 2.24$ and 2.0, respectively (full lines) compared with experiment (Ref. 1; $\Box$ magnetic field $H \parallel$ c, $\bigcirc$ $H \perp$ (a,b). In the inset the DC scenario (a) is compared with the single-chain one (b). Thickness of lines symbolizes the coupling strength. The empirical $J$-values are in accord with LDA and microscopic estimates.](https://example.com/figure1.png)
mation, e.g. as in Li$_2$CuO$_2$ with fm inchain order below 8K.