Comment on “Two-spinon and four-spinon continuum in a frustrated ferromagnetic spin-1/2 chain” by M. Enderle et al.

In a recent inelastic neutron scattering (INS) study of LiCuVO$_4$ [1] a measurement of the dynamic spin susceptibility Im$\chi$($\omega$, $q$) has been reported. The authors claim that (i) it is well described by two weakly ferromagnetically (FM) coupled interpenetrating Heisenberg antiferromagnetic (AFM) spin-1/2 chains (HAF), (ii) the extracted exchange integrals $J_i$ agree with a spin-wave description [2], (iii) the INS intensity above 10 meV has been ascribed to a 4-spinon continuum. Their $J$-set reads $J_1=-1.6$ meV, $J_2=3.56$ meV for the NN and NNN in-chain couplings, respectively, (i.e. $\alpha=-J_2/J_1=2.25$) and $J_{\perp}=-0.4$ meV ($J_3$ in Ref. 1) for the diagonal interchain coupling in the (ab)-plane.

We will show that LiVCuO$_4$ should be described by strongly ferromagnetically coupled HAF’s and for the $J$’s given in Ref. 1 all issues (i)-(iii) do not hold. An alternative set in accord with various experimental results, including the INS data, and findings of independent theoretical studies is proposed. In view of the recent possible discovery of quantum-spin nematics and Bose condensation of two-magnon bound states [3, 4] in LiCuVO$_4$ a precise knowledge of the main $J$’s is of key importance.

We start with the high-temperature (HT) spin susceptibility $\chi(T)$ shown in Fig. 1a (see Fig. 4 in Ref. 2). From a linear fit of these data for 500 K $\leq$ $T$ $\leq$ 650 K, we arrive at a FM Curie-Weiss temperature $\Theta_{\text{CW}} = +7.4$ K. For weak $J$’s it excludes a dominant AFM $J_2$ value. Introducing $\theta = -2\Theta_{\text{CW}}/J_1$, $j_{\perp} = -J_{\perp}/J_1$ one obtains for the 2D spin-model under consideration an exact constraint:

$$1 = \alpha + \theta + 2j_{\perp}. \quad (1)$$

For the $J$’s from Ref. 1 the right hand side of Eq. (1) yields 2.52, i.e. a clear violation. Only for an unphysical value of $\theta < -1$ a large $\alpha > 2$ could be compensated. Note that values $\theta < 0$ have been reported [2, 4], but they are based inappropriately on fits at still too low $T$ (see Fig. 1a). In contrast for a microscopically derived set (see below) $J_1 = -6.3$ meV and $\alpha \approx 0.8$, $\theta = +0.2$, and a tiny $j_{\perp}$ Eq. (1) is obeyed. The inspection of Fig. 1(b,c) shows that the dispersion of the INS main peaks (red curves) is not enough to find a unique $J_1$-$J_2$ set. The INS spectral density must be considered, too. Our set explains also the larger INS intensity above 9.5 meV at variance to almost no intensity for the set given in Refs. 1,2 (compare the boxes in Figs. 1(b,c)).

A mapping from a Cu3$d$ O2$p$ five-band Hubbard model with usual parameters which describes the $T$-dependent dielectric response [4] onto a $J_1$-$J_2$ spin-1/2 model yields a sizeable $J_1 = -6.3$ meV and $J_2 = 5.05$ meV. We stress that in all closely related sister compounds with a Cu-O-Cu bond angle $\sim$95° FM $|J_1|$ values $\approx$ 1.6 meV have been found in fitting various data: Li$_2$CuO$_2$: $J_1 = -19.6$ meV (INS [4]), Ca$_2$Y$_2$Cu$_3$O$_{10}$: $-14.7$ meV (INS [5]), Li$_2$ZrCuO$_4$: $-23.7$ meV ($\chi(T)$, $c_p$ [10]) (11). We have performed also total energy calculations within the LSDA+$U$ for various magnetic structures [6] and arrived with $U=6\pm0.5$ eV at $J_1 = -8.8\pm0.3$ meV, $J_2 = 6.5 \pm 1$ meV, and $J_{\perp} = 0.5 \pm 0.05$ meV ($J_4$ in Ref. 2).

To conclude, the application of a $J_1$-$J_2$ model with a weak FM $J_1$ ($\alpha > 2$) to LiCuVO$_4$ as in Ref. 1 is not justified whereas a strong coupling regime ($\alpha < 1$) with comparable and sizeable FM $J_1$ and AFM $J_2$ exceeding much the $J$’s given in Ref. 1 is consistent with the INS data and the physics of edge-shared CuO$_2$ chains. For strongly coupled HAF’s a separation of 2- from 4-spinon contributions is nontrivial and requires special theoretical investigation. A straightforward assignment of spectral features to a 4-spinon continuum valid only in the opposite limit $\alpha > 1$ is therefore precluded.

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$^1$ Inst. f. Theor. Solid State Physics, IFW-Dresden 01171 Dresden, Germany, email: drechsler@ifw-dresden.de
$^2$ Institute of Physics, ASCR, Prague, Czech Republic
$^4$ MPI-CPS Dresden, Germany.