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 $\mathrm{Im}\chi(\omega, q)$ 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. $\alpha=-J_2/J_1$=2.225) and $J_{\rm ic}$=−0.4 meV (J_5 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 $\Theta_{\rm cw}$ = +7.4 K. For weak $J_{\rm ic}$ it excludes a dominant AFM J_2 value. Introducing $\theta = -2\Theta_{\rm cw}/J_1$, $j_{\rm ic} = -J_{\rm ic}/J_1$ one obtains for the 2D spin-model under consideration an exact constraint:

$$1 = \alpha + \theta + 2j_{\rm ic}. \qquad (1)$$

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 $\alpha \approx 0.8$, θ=+0.2, and a tiny $j_{\rm ic}$ 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)).

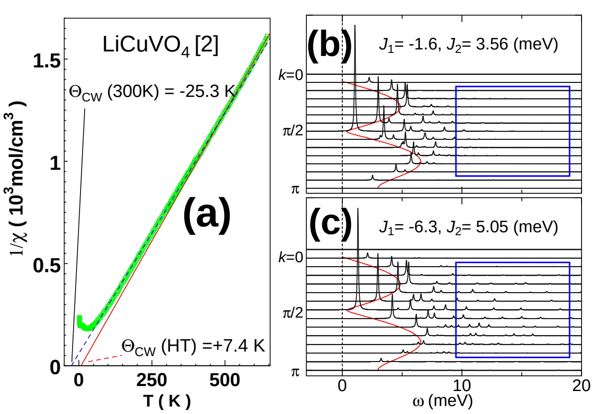

FIG. 1: (Color) Inverse spin susceptibility of $LiCuVO_4$ vs. temperature (a). Dynamical structure factor from exact diagonalizations for a single chain with N=28 sites broadened with 0.05 meV for the J-set of Ref. 1 (b) and for our set (c). Red curves: spin-wave description of INS peaks [2].

A mapping from a Cu3d O2p five-band Hubbard model with usual parameters which describes the T-dependent dielectric response [6, 7] 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 $\lesssim$95° FM $|J_1|$-values $\gg$ 1.6 meV have been found in fitting various data: Li_2CuO_2: J_1=−19.6 meV (INS [8]), $Ca_2Y_2Cu_5O_{10}$:−14.7 meV (INS [9]), Li_2ZrCuO_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 [5] and arrived with U=6±0.5 eV at J_1=−8.8±0.3 meV, J_2=6.5±1 meV, and $J_{{\rm ic}\,\perp}$=0.5±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 (α >2) to $LiCuVO_4$ as in Ref. 1 is not justified whereas a strong coupling regime (α <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 \gg$1 is therefore precluded.

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