The strength of frustration and quantum fluctuations in LiVCuO$_4$

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Abstract – For the 1D-frustrated ferromagnetic $J_1$-$J_2$ model with inter-chain coupling added, we analyze the dynamical and static spin structure factor $S(k, \omega)$, the pitch angle $\phi$ of the magnetic structure, the magnetization curve of edge-shared chain cuprates, and focus on LiCuVO$_4$ for which neither a perturbed spin one nor a spin wave approach can be applied. $\phi$ is found to be most sensitive to the interplay of frustration and quantum fluctuations. For LiCuVO$_4$ the obtained $J$’s agree with the results for a realistic 5-band extended Hubbard model and LSDA + $U$ predictions yielding $\alpha = J_2/|J_1| \approx 0.75$ in contrast to $5.5 > \alpha > 1.42$ suggested in the literature. The $\alpha$-regime of the empirical $\phi$-values in NaCu$_2$O$_2$ and linarite are considered, too.

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Introduction. – LiCuVO$_4$ is one of the first [1,2], but still often studied spin-chain compounds among edge-shared cuprates (ESC) [3–7]. Special interest is caused by its multiferroicity [8–10], by a possible realization of spin nematics and related Bose-Einstein condensation of two-magnon bound states in high magnetic fields [11–13]. Both phenomena are not fully understood and a precise knowledge of the main exchange integrals is of key importance to attack such complex problems in a realistic way. So far, rather different values of the in-chain frustration $\alpha = J_2/|J_1|$ with $0.5 \leq \alpha \leq 2.2$ [3,5,14–16] and recently even above 5.5 [17] have been reported. Here, $J_1 < 0$ is the ferromagnetic (FM) nearest neighbor (NN) and $J_2$ the antiferromagnetic (AFM) next-nearest neighbor (NNN) coupling in chain direction $b$ (fig. 1). The scatter of $\alpha$ stems from the fact that, despite a common misconception, any spectroscopy does not measure the interaction constants directly. Instead, it measures observables (e.g., the spin structure factor (SSF) in case of inelastic neutron scattering (INS)). Their relation to the exchange constants is non-universal and model dependent. Keeping in mind the weak inter-chain coupling (IC), the single-chain can be viewed also as two interacting and interpenetrating AFM Heisenberg chains (AHC) or equivalently as a single zigzag ladder. Then, one is left with a weak- ($\alpha > 1$) or a strong-coupling scenario ($0 < \alpha < 1$). $x \equiv 1/\alpha$ provides a direct measure of the FM “IC” between the legs of a zigzag ladder (i.e., the interpenetrating AHC). Here, we show...
that a proper handling of strong quantum fluctuations (QF) unambiguously evidences \( \alpha < 1 \) which leads to a substantially different parameter assignment as compared to refs. [3,5] and [17]. Strong QF manifest themselves by a small ordered magnetic moment and a low Néel temperature [2,10]. Here we elaborate the approaches outlined in ref. [15] and critically discuss in more detail also arguments put forward in ref. [6] to justify a weak-coupling scenario. Strong QF addressed here are also relevant for the ESC NaCu2O2 but ignored in its \( \alpha \)-assignment [18].

**Phenomenological analysis.** – In spite of the \( k \)-dependent Cu\(^{2+}\) form factor, the intensity of the INS spectrum is proportional to the dynamic SSF \( S(k, \omega) \). In quasi-2D and 3D magnets, at \( T \) well below the ordering temperatures, magnons are almost undamped magnetic excitations, then \( S(k, \omega) \propto \delta(\omega - \omega_k) \), and the dispersion \( \omega_k \) provides the exchange interactions. In isolated frustrated \( J_1-J_2 \) 1D spin chains magnons are undamped excitations above a FM ground state for \( 4 < x < +\infty \), and \( \omega_k \) unambiguously determines \( J_1 \) and \( J_2 \) [19]. The relation between \( J_2 \) and the SSF is also known for \( x = 0 \), when the chain under consideration is given by two interpenetrating but noninteracting AHC. In this limit, magnons decay into pairs of spinons, the SSF is non-zero for \( \omega > \omega_{\text{L},k} = (\pi/2)J_2\sin(2k) \), and the INS spectrum is symmetric for \( \pi/2 \pm k \). But for \( 0 < 1/\alpha < 4 \) relevant here, no exact relation between the SSF and \( J_1 \), \( J_2 \) is available. For real quasi-1D systems, the problem is more involved. In the case of coupled AHC, the IC confines spinons and restores the magnon peaks in the SSF at sufficiently low energies and \( T \) [20]. The dispersion of these peaks may be described in terms of spin wave theory (SWT) with some effective \( J^\text{SWT} \)-s. The relation to the Hamiltonian parameters can be highly non-trivial [21]. Such a fit for LiVCuO4 has been reported also in ref. [3]. It is sketched by the red curves in fig. 2(a)–(c) and the obtained parameter set is shown in table 1 together with the \textit{ad hoc} renormalization (AR) by the factor \( 2/\pi \) only for AFM couplings proposed therein. Based on these and recent high-energy data (20 meV > \( \omega > 0.5 \) meV data) analyzed by means of a random phase approximation (RPA) for the account of the coupling between the AHC via an \textit{unrenormalized} \( J_1 \) within an effective 1D model has been proposed in ref. [5] (see table 1):

\[
J_{\text{eff},2} \approx (2/\pi)J_2^{\text{SWT}} \quad \text{and} \quad J_{\text{eff},1} \approx J_1^{\text{SWT}} + 2J_2^{\text{SWT}}.
\]

(1)

If the IC is of less relevance for the “high-energy” physics, the claimed \( 2J_2 \approx -0.8 \) meV should be added to \( J_2^{\text{RPA}} = -2.4 \) meV only for low-energy problems such as thermodynamics, \( i.e., \) relevant for the saturation field and the magnetization or the determination of the spiral’s pitch angle \( \phi \). With such a more convincing empirical RPA affected renormalization one would already arrive at \( \alpha = 1.063 \) close to the strong-coupling border line. Up to now all considerations were based on the assumption that the FM \( J_1 \) remains fixed. However, field theory flow-equations based approaches [22] valid at \( x \ll 1 \) point to strong-coupling renormalizations. As a consequence, \( J_1 \) might change considerably and \( \alpha \) is further scaled down. Our results obtained using the density matrix renormalization group (DMRG) technique [23] (see fig. 2 and table 1) confirm this tendency [15]. If one adopts the AR set as a starting point, our results demonstrate upward renormalizations: strong for \( |J_1| \) and moderate for \( J_2 \).

We start our analysis of available INS data [3,5] with the two maxima of the lowest peak dispersion \( \Omega_\pm \approx 4.84 \) meV and \( \Omega_\pm \approx 6.4 \) meV near the transferred momenta \( k = 1/4 \) and \( k = 3/4 \). Although the maximum corresponding to \( \Omega_+ \) is broad, the asymmetry with respect to \( \Omega_- \) quantified by our dynamical asymmetry parameter \( \rho = \Omega_+/\Omega_- \approx 1.32 \) is clearly visible in the INS-data in contrast to the set proposed in ref. [17] (see table 1) where \( \rho \approx 1 \) would occur. On the absolute scale a deviation by a factor > 3 between the INS [5] and the predicted dispersion is observed (see fig. 2(c)) which can be traced back to the anomalously large values of \( J_2 \) obtained in ref. [17].

The dependence of \( \rho \) on \( x = 1/\alpha \) can be obtained from fitting our dynamical DMRG [24] results for \( 0.3 \leq \alpha \leq 3 \) and long chains with \( L = 96 \) sites

\[
\begin{align*}
\Omega_+ & = \frac{\pi}{2} + 0.0338x - 0.302x^2 + 0.0831x^3 - 0.00699x^4, \\
\Omega_- & = \frac{\pi}{2} - 0.143x - 0.534x^2 + 0.279x^3 - 0.0589x^4 \\
& + 0.0046x^5.
\end{align*}
\]

(2)

The function \( \rho(x) \) provides a convenient sensitive measure of the interaction regime which is heavily affected by the strong QF. This function is depicted in fig. 2(h). One realizes excellent agreement with \( \alpha = 0.75 \) derived in our previous paper where instead of \( \rho \), \( \Omega_- \) and the relative magnetization curve \( M(H/H_0)/M_s \) at low temperature have been employed [15]. Notice the large deviations in \( x \), if the SWT, AR or the RPA would be applied to extract its value (see fig. 2(h)). The strong deviations of both values from our DMRG-based value clearly show the inapplicability of simple spin wave theory based estimates. The physical reason is the neglect of strong QF in LiVCuO4. Strong QF manifest themselves also in its small magnetic moment as mentioned above and in relatively large \( \phi \)-values (see below).

Finally, considering briefly the calculated and the experimental INS intensities, at present only few comparisons are possible due to unpublished complete INS spectra. Nevertheless, comparing, \( e.g., \) the available data shown in fig. 2(e) with our dynamical DMRG results shown in figs. 2(f), (g) one realizes that our set provides a much better description of the INS intensity at large transferred momenta \( k > \pi/2 \) as compared with that of ref. [5].

If one adopts that the experimental magnetization data up to the so-called field \( H_{\text{d}} \approx 40.5 \pm 0.2 \) T (\( H \parallel c \)) where the main peak in \( \mathrm{d}M/\mathrm{d}H \) occurs [12] is well described by an effective 1D model, one arrives at the curves shown in
Fig. 2: Left: magnetic dynamical structure factor $S(k,\omega)$ from exact diagonalizations for periodic chains and $L=28$ sites for the $J$-sets proposed in ref. [5] (a), in our recent work [15] (b), and in ref. [17] for $U=5\epsilon$ (see table 1) (c). Right: experimental asymmetric dispersion along the chains in between $\Omega_+$ and $\Omega_-$ (see panel (b) and eq. (2)) and INS-intensities from ref. [5] ((d), (e)), calculated $S(k,\omega)$ normalized to the static structure factor $S(k)$ (i.e., the $\omega$-integrated $S(k,\omega)$) for a single chain with our parameters [15] (f) and that of ref. [5] (g), and the ratio of the peak positions at the 1st and 2nd maximum of $S(k,\omega)$ for the transferred momenta near $k=1/4$ and $3/4$, respectively, (h) given by the red curves in (a)–(c). In all DMRG-calculations for open chains and $L = 96$ sites a Lorentz broadening at half-width $\Gamma = 0.3\,$meV has been used.

Fig. 3. Notice the strong deviation of the weak-coupling set from in ref. [17].

Then, a dominant FM IC as proposed in refs. [3] and [5] cannot be reconciled with these experimental data since for such a coupling (see table 1) the 3D saturation field is smaller than its 1D counterpart [25]. Also the spin susceptibility $\chi(T)$ is within an RPA approach for the inter-chain coupling best described by a total AFM IC. Anyhow, a detailed discussion of $\chi_\alpha(T)$ including also the background susceptibility $\chi_0$ will be given elsewhere.

The presence of strong QF is evidenced by the small magnetic moment of $0.31\mu_B$ and a low Neél temperature $T_N = 2.4\,$K. Both values should be compared, e.g., with the much larger values $0.97\mu_B$ and $9\,$K for the sister compound Li$_2$CuO$_2$ [26,27] caused by a relatively strong IC [28] and a small $\alpha = 0.32$ which also reduces the strong QF. Although the magnetic moments of the ESC linarite and NaCu$_2$O$_2$, $0.5\mu_B$ and $0.73\mu_B$ [29], respectively, exceed that of LiVCuO$_4$, the presence of strong QF is still obvious. Hence, the spiral state of LiVCuO$_4$ is even
Table 1: Exchange integrals (in meV) from INS data using bare spin wave theory (SWT), ad hoc renormalizations (AR) [3] or RPA [5] as compared with sets derived from microscopic models (see below). In all LDA(GGA) + U calculations shown a value \( U_{3d} - J_{3d} = 5 \text{ eV} \) appropriate for ESC has been used.

|          | \( J_1 \) | \( J_2 \) | \( \alpha \) | \( J_4 \) | \( J_5 \) |
|----------|-----------|-----------|-------------|-----------|-----------|
| Bare SWT [3] | -1.6      | 5.59      | 3.49        | 0.01      | -0.4      |
| AR [3]    | -1.6      | 3.56      | 2.23        |           |           |
| RPA [5]   | -2.4      | 3.4       | 1.42        |           |           |
| Present work | -6.95     | 5.2       | 0.75        |           |           |
| 3dO2p optics | -6.31     | 5.05      | 0.8         |           |           |
| GGA + U [17] | -2.7      | 15.0     | 5.55        | -1.31     | 0.16      |
| LSDA + U   | -8.5      | 7.05      | 0.82        |           |           |
| GGA + U   | -6.4      | 5.45      | 0.85        |           |           |

Fig. 3: Magnetization vs. applied field. Experiment (green circles) from ref. [12], theory: blue line - set of refs. [5], red line - our set [15] and black line - the \( U = 5 \text{ eV} \) set from ref. [17] and \( g = 2.3 \) for all sets. The DMRG calculations were performed for \( L = 512 \) sites at \( T = 0 \). Inset: the “entire” field range.

more strongly driven towards decoupled AHC with a corresponding collinear Neél state, for each leg. Thus, the experimental pitch \( \phi = 84^\circ \) analyzed within the SWT or AR approaches results in strongly overestimated \( \alpha \)-values. This is illustrated in figs. 4 and 5, where the maximum of the static magnetic structure factor is depicted as a function of \( \alpha \) for the cases of a frustrated \( J_1-J_2 \) single chain and a coupled pair of them, respectively. Already the latter is expected to provide insight into the real quasi-1D situation. This point of view is supported by coupled cluster calculations to be reported elsewhere. Thus, for instance in case of a dominant 2D IC as in the model adopted in refs. [3] and [10–12], the effective IC \( J_5^* \) and \( J_4^* \) read

\[
J_5^* = 2J_5, \quad J_4^* = 2J_4.
\]

Notice the failure of the classical curve especially for not too large \( \alpha \) (see fig. 4). Such an effect was first addressed in 1D by means of DMRG [30] and in ref. [31] for a plane of perpendicularly coupled chains using a coupled cluster approach. We stress that the experimental pitch \( \phi = 84^\circ \) [2] is reproduced for \( \alpha \leq 1 \), only, irrespective of the details of the weak IC. Adopting the in-chain \( J \)'s and the leading IC \( J_5 \approx -0.4 \text{ meV} \) suggested in ref. [3], one estimates from fig. 5 a pitch angle of 89.58° (\( \alpha = 2.22 \)) and
of 89.43° in the case of the RPA set (α = 1.42) in contrast to 84° known experimentally [2]. Thus, the measured φ points clearly to a strong-coupling regime in contrast to statements in refs. [3,5] and [17]. Naturally, since the SWT derived J’s obey the classical relation

\[ \phi_{cl} = \cos^{-1}\left( -\frac{J_1 + 2J_2}{4J_2} \right) \]  

valid in case of an in-phase ordering of spirals in the (a-b)-plane, they cannot be reconciled also with strong QE. Equation (4) yields 84.0° for α = 3.5 [3] and J_5 = −0.37 meV [3] (the small deviation from 84.24° corresponding to the observed propagation vector results from other weak IC J_5, J_4, and J_0 ignored in eq. (4) for the sake of simplicity) and 87.42° for α = 5.55 [17]. What matters here is not the value of φ itself, but the difference π/2 − φ which differs by an order of magnitude between the quantum and the classic case [32]. Hence, it is convenient to measure the role of QE for a given set of J’s by the expression

\[ \nu = \frac{\pi/2 - \phi_{cl}}{\pi/2 - \phi} - 1 = \frac{\phi - \phi_{cl}}{\pi/2 - \phi} > 0. \]  

The 1D and the quasi-1D (2D) results are shown in fig. 6. Just in the region α ~ 1 to 2 the quantum effect (QE) is most pronounced, whereas near the critical α_c = 0.25 and in the extreme weak-coupling limit 1/α ≪ 1 the QE for φ disappears. Thus, in this limit the QE for φ should be distinguished from the strong renormalization of the dispersion of spin excitation (see eq. (1)). Thus, the attempt to describe the spin dynamics in the intermediate coupling regime quasi-classically is in our opinion the main reason for the rather different α-assignment in ref. [3]. A similar unconvincing assignment α = 5.5 and J_1 ≈ −1.4 meV, only, but with long-range in-chain J_3 = 0.63 meV and J_4 = 0.54 meV, has been reported for NaCu_2O_2 [18] from fitting the observed pitch φ = 82° by a classical expression like eq. (4). The reported α = 1.6 for linarit [33] is in conflict with the observed pitch φ ≈ 34° [29,33] and α ~ 0.36 [34].

**Microscopic analysis.** – Turning to a microscopic analysis, we compare our (DMRG) INS derived J’s with those from two independent approaches: i) analyzing high-energy spectra from EELS, optical conductivity \( \sigma(\omega) \) or resonant inelastic X-ray spectroscopy (RIXS) data within a strongly correlated extended multiband Hubbard model and a subsequent mapping of its spin states onto the corresponding states of the 1D J_1-J_2 spin model under consideration (see table 1); ii) extracting these J’s from total energy calculations of different prepared magnetically ordered states (see, e.g., [17], table 1, and fig. 7). Mapping an extended Cu_3dO_2p five-band Hubbard model (ESBHIM) with usual parameters which describes the T-dependent dielectric response [35,36] onto a J_1-J_2 spin-(1/2) model yields also a sizeable J_1 = −6.3 meV and J_2 = 5.05 meV, similarly to all closely related sister ESC [37] with a Cu-O-Cu bond angle ≲ 95°. Here sizeable FM J_1- values >> 1.6 meV have been found in fitting various data: Li_2CuO_2: J_1 = −19.6 meV (INS [28]), Ca_2Y_2Cu_3O_10: J_1 = −14.7 meV (INS [19]), Li_2ZrCuO_4: J_1 = −23.7 meV (\( \chi(\Omega) \), \( \sigma_p \) [14,38]), and linarite: J_1 = −11.9 meV M(\( H \)), \( H_{sat} \) [34]. In particular, also for LiVCuO_4 the T-dependent optical conductivity data [35] can be well fitted within an ESBHM on chain-clusters with up to six edge-shared CuO plaquettes. Thereby \( U_d = 8 \) eV, \( U_p = 4.1 \) eV \( K_{pd} = 65 \) meV, \( \Delta_{pd} = 3.82 \) eV etc. has been used. Then one arrives at in-chain J’s close to the INS-derived ones: α = 0.8 and J_2 = 5.1 meV (see table 1). The value of J_1 is sensitive to the magnitude of the direct FM exchange \( K_{pd} \), whereas J_2 is mainly sensitive to the in-chain O-O transfer integrals. Thereby \( |J_1| \propto K_{pd} \) holds approximately. Notice that \( K_{pd} \) is much more important for the large value of −J_1 than the FM Hund rule coupling on O. In the past \( K_{pd} \) has been used mostly as a fitting parameter ranging from 50 to 110 meV for CuGeO_3 [39,40]. A reliable J_1-value derived from an INS analysis as reported here is helpful to restrict its value and opens a door for systematic studies of this important microscopic exchange and useful comparisons with other ESC and cuprates in general [37]. In fact, our empirical \( K_{pd}-\)value corresponds to 130 meV for a \( \sigma\)-Cu-O bond as compared with 180 meV for a CuO_2 plane [41].

The main J’s can be also extracted from the total energies of various magnetic states calculated within the local density approximation (LDA + U) or within the generalized gradient one (GGA + U). Thereby the results depend mainly on a single parameter \( U = U_{3d} - J_{3d} \), where \( J_{3d} \approx 1 \) eV denotes Hund’s rule coupling that is rather precisely known for transition metals. For both approximations, we calculated J_1 and J_2 for the two crystal structures refined from X-ray diffraction and neutron diffraction (labeled XRD and ND, respectively). The resulting α
Fig. 7: Frustration parameter $\alpha$ vs. effective on-site repulsion $U$ for different crystal structure refinements from X-ray (XRD) and neutron diffraction (ND). Inset: the obtained $\alpha$-values vs. $U$ compared with the results of ref. [17].

is shown in fig. 7 and in table 1 (for $U = 5 \text{ eV}$). The graph indicates only small differences for the two crystal structure solutions, but essentially no difference for the two choices of the exchange-correlation potential (LDA + $U$ vs. GGA + $U$). For realistic $U$’s which describe successfully other ESC, one arrives again at $\alpha < 1$ in contrast to ref. [17], which reports unusually large $J_2$- and $\alpha$-values incompatible with the observed $\phi$ [2], the restricted two-spinor continuum, and an obviously asymmetric INS spectrum [5]. Presumably it is a consequence of the double counting procedure employed in ref. [17] and not an artifact of the GGA in general as stated there since our calculations shown in fig. 7 yield close values in the $\alpha$-region of interest, both for the LDA and the GGA. The RPA-derived value $\alpha_{\text{RPA}} \sim 1.4$ [5] could be only approached for unrealistically small $U$-values $< 3 \text{ eV}$ adopting the XRD data.

Summary. – A revisited analysis of LiVCuO$_4$ provides clear evidence for strong coupling in terms of antiferromagnetic Heisenberg chains. It is based on a careful revised interpretation of experimental data. The dynamical asymmetry parameter $\rho$ given by INS, and the pitch angle are very sensitive to quantum fluctuations and frustration. Weak coupling ($\alpha > 1$) would result in a nearly collinear leg state ($< 1^\circ$ deviations from the 90$^\circ$ pitch limit of decoupled legs) and in almost vanishing dynamical anisotropy ($\rho \to 1$) incompatible with diffraction and INS data. Our obtained main $J$-values are supported by independent microscopic studies based on the L(S)DA + $U$ approach and the extended Cu3dO2p Hubbard model.

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