Microscopic model for Bose-Einstein condensation and quasiparticle decay

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**Abstract** – Sufficiently dimerized quantum antiferromagnets display elementary $S=1$ excitations, triplon quasiparticles, protected by a gap at low energies. At higher energies, the triplons may decay into two or more triplons. A strong enough magnetic field induces Bose-Einstein condensation of triplons. For both phenomena the compound IPA-CuCl$_3$ is an excellent model system. Nevertheless no quantitative model was determined so far despite numerous studies. Recent theoretical progress allows us to analyse data of inelastic neutron scattering (INS) and of magnetic susceptibility to determine the four magnetic couplings $J_1 \approx -2.3$ meV, $J_2 \approx 1.2$ meV, $J_3 \approx 2.9$ meV and $J_4 \approx -0.3$ meV. These couplings determine IPA-CuCl$_3$ as system of coupled asymmetric $S=1/2$ Heisenberg ladders quantitatively. The magnetic field dependence of the lowest modes in the condensed phase as well as the temperature dependence of the gap without magnetic field corroborate this microscopic model.

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Low-dimensional antiferromagnetic quantum spin systems display various fascinating properties, e.g., spin-Peierls transition [1,2], appearance of a Haldane gap for integer spins [3,4], high-temperature superconductivity upon doping [5], and the Bose-Einstein condensation (BEC) in spin-dimer systems [6–9], where the latter one is characterized by a phase transition from a non-magnetic phase to a long-range antiferromagnetically ordered gapless phase at a critical magnetic field $H_c$.

Another fascinating phenomenon recently observed in low-dimensional antiferromagnets is the decay of their elementary $S=1$ excitations, triplons [10], at higher energies so that the triplons exist only in a restricted part of the Brillouin zone [11,12]. Theoretically as well, there is rising interest in the understanding and quantitative description of this phenomenon for gapped triplons [13–16] as well as for gapless magnons [17–19].

The description of quasiparticle decay faces an intrinsic difficulty. The merging of the long-lived, infinitely sharp elementary triplon with a multitriplon continuum requires to describe the resulting resonance and its edges precisely. This is still a challenge for numerical approaches such as exact diagonalization or dynamic density-matrix renormalization [20]. Diagrammatic approaches are able to capture the qualitative features but may encounter difficulties in the quantitative description in the regime of strong merging where the sharp mode dissolves completely in the continuum because this is a strong-coupling phenomenon [13,14]. Unitary transformations also face difficulties when modes of finite lifetime occur [16].

A crucial step in the understanding of both phenomena is to identify a suitable experimental system. The best studied candidate for the BEC in coupled spin-dimer systems is TiCuCl$_3$. Unfortunately, recent research suggests that the high-field spectrum remains gapped [21,22] in contrast to what is expected from a phase where a continuous symmetry is broken. This suggests the existence of anisotropies. A promising alternative for a BEC in a spin-dimer system is (CH$_3$)$_2$CHNH$_3$CuCl$_3$ (isopropylammonium trichlorocuprate(II), short: IPA-CuCl$_3$) where inelastic neutron scattering (INS) provides evidence for an almost exact realization of a BEC [9,23].

A suitable experimental system to study triplon decay in detail is searched for. The two-dimensional (2D) PHCC [11,24] is a candidate, but it involves eight different couplings so that a quantitative characterization is impossible to date. Due to its quasi–one-dimensional

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(quasi-1D) structure, IPA-CuCl$_3$ is again a more promising candidate. This compound seems to realize the theoretically proposed situation for BEC in coupled spin ladders [8].

But in spite of many years of intensive studies [9,12,25–29] no quantitative microscopic model for IPA-CuCl$_3$ is established. The present work aims at filling this gap. Theoretically, our study is based on continuous unitary transformations (CUTs) of models with quasiparticle decay [16] and on high-temperature series expansions for asymmetric spin ladders which are topologically equivalent to dimerized and frustrated spin chains [30]. The experimental input used in INS data [12] and magnetic susceptibility $\chi(T)$ data [26]. We will illustrate why it is intrinsically difficult to determine the microscopic model.

Finally, we will compute the temperature and the magnetic field dependence of the lowest magnetic modes as well as the upper critical magnetic field $H_{c,2}$, which induces full polarization. They all agree very well with experimental data [9,23,31–33] which supports the advocated model.

Since the characterization of IPA-CuCl$_3$ by Roberts et al. [25] various spin models were discussed. Manaka et al. pointed out that the magnetic susceptibility of IPA-CuCl$_3$ can be explained by a ferro-antiferromagnetically alternating Heisenberg $S=1/2$ chain with ferromagnetic coupling twice as large as the antiferromagnetic coupling [26]. According to Hida [34] the magnetic ground state is thus given by a gapped Haldane state [3].

The dispersions measured by INS [12] and the crystal structure of IPA-CuCl$_3$ indicates that the system is quasi-2D. It is described by weakly coupled asymmetric spin $S=1/2$ Heisenberg ladders, see fig. 1, with

$$H = H_{1D} + H_{\perp},$$

$$H_{1D} = J_1 \sum_{r,s} S_{1,r,s} S_{2,r+1,s} + J_3 \sum_{r,s} S_{1,r,s} S_{2,\overline{r},s} + J_2 \sum_{r,s} (S_{1,r,s} S_{1,r+1,s} + S_{2,r,s} S_{2,r+1,s}),$$

$$H_{\perp} = J_4 \sum_{r,s} S_{1,r,s} S_{2,r+1,s+1},$$

with two ferromagnetic couplings $J_1$, $J_4 < 0$ and two antiferromagnetic couplings $J_2$, $J_3 > 0$. The dominant dimer coupling is $J_4$ so that we use the ratios $x = J_2/J_3$, $y = J_1/J_3$ and $z = J_4/J_3$. Let us first consider the ladders as isolated because the interladder coupling is small. The standard view of these ladders takes the $J_3$ bonds to form the rungs of the ladder. Then $J_1$ is a diagonal bond.

The key element of this model is the asymmetry of the spin ladders controlled by $J_1$. On the one hand, the presence of $J_1$ spoils the reflection symmetry about the center line of the ladder between the legs. This asymmetry would imply a conserved parity such that the triplons on the dimers could be changed only by an even number [35,36] so that no decay of a triplon into a pair of triplons could occur. Hence the very presence of $J_1$ opens an important decay channel for quasiparticle decay.

On the other hand, the two bonds $J_2$ and $J_1$ represent the coupling of adjacent dimers. Both contribute to the hopping of the triplons which is given in leading order by $2J_2 - J_1$ [37], while the interaction of adjacent triplons is proportional to $2J_2 + J_1$. With information only on the dispersion [12] it is impossible to determine $J_1$ and $J_2$ separately. Hence, the same feature that induces the interesting quasiparticle decay makes it particularly difficult to establish a microscopic model.

The BEC occurring in TlCuCl$_3$ was successfully described by the bond-operator approach [38,39]. But this approach to spin-dimer systems is quantitatively reliable only as long as the interdimer couplings $J_{\text{inter}}$ are significantly smaller than the dimer coupling $J_{\text{dimer}}$: $|J_{\text{inter}}| < J_{\text{dimer}}/2$ [40]. This limit requires $|J_1| < J_3/2$ for $i \in \{1,2,4\}$ for IPA-CuCl$_3$ which does not hold [26]. We will see below that $|J_1| \approx J_3$ provides very good fits.

Thus we apply self-similar CUTs (sCUTs) to isolated ladders [41–44], modified to cope with decaying quasiparticles [16]. We use an infinitesimal generator which decouples the subspaces with zero or one triplons from the remaining Hilbert space. We can still decouple the 1-triplon subspace from the 2-triplon subspace for the isolated ladder. The proliferating flow equations are truncated if the range of the corresponding process exceeds certain maximum extensions in real space. Thereby, the ladders are mapped to an effective model

$$H_{1D,\text{eff}} = \sum_{h,l,a} \omega_h^a(h) t^a_{h,l} t^a_{h,l}$$

1The truncation scheme used for the Hamiltonian is $(d_0, d_1, \ldots, d_6) = (10, 8, 8, 5, 3, 3)$ and for the observables $(d_1, d_2, \ldots, d_6) = (10, 10, 8, 8, 6, 6)$, where $d_i$ is the maximum extension for a process with $j$ creation and annihilation operators. Additionally, we keep only terms that create or annihilate at most $N=4$ triplons in the Hamiltonian and $N=3$ triplons in the observables, see also refs. [16,49].
in terms of triplon creation $t^\dagger_{\alpha,h,l}$ and annihilation operators $t_{\alpha,h,l}$ in momentum space, where $h$ is the wave vector component along the ladders, $l$ the one perpendicular to them, and $\alpha \in \{x, y, z\}$ the spin polarization. These operators are the Fourier transforms of the bond operators [45,46] defined on the dimers in fig. 1.

The dispersion $\omega(h)$ depends only on $h$ because the CUT is applied to the isolated ladders which still have to be coupled. This coupling is achieved in leading order following the approach in refs. [47,48]. The spin component $S^{\alpha}_{i,r,s}$ is taken as observable and transformed into the new basis by the CUT. Then it reads

$$S^{\alpha}_{\text{eff},i,r,s} = U^{\dagger}S^{\alpha}_{i,r,s}U \quad \text{(3)}$$

where the coefficients $a_{i,\delta}$ result from the CUT, cf. refs. [47,48]; the dots stand for normal-ordered higher terms in the real space triplon operators $t^\dagger_{\alpha,h,l}(t_{\alpha,h,l})$. Knowing $S^{\alpha}_{\text{eff},i,r,s}$ allows us in a second step to write down the effective interladder coupling $H_{\text{int,eff}}$ in real space

$$H_{\text{int,eff}} = J_4 \sum_{r,s,\alpha,\delta,\delta'} a_{r,\delta} a_{s,\delta'} [t^\dagger_{\alpha,r,s}(t^\dagger_{\alpha,r+1+(\delta'-\delta),s+1} + t_{\alpha,r+1+(\delta'-\delta),s+1}) + \text{H.c.}] \quad \text{(4)}$$

This neglects trilinear and higher contributions. The Fourier transform of $H_{\text{int,eff}}$ leads to $H_{\text{eff}}=H_{\text{1D,eff}}+H_{\text{int,eff}}$ amenable to a Bogoliubov diagonalization if the hardcore property of the triplons neglected. This yields

$$H_{\text{eff}} = \sum_{h,l,\alpha} \omega(h,l) \tau^\dagger_{\alpha,h,l} \tau_{\alpha,h,l} \quad \text{(5a)}$$

$$\omega(h,l) = \sqrt{\omega_0^2(h) + 4\omega_0(h)\lambda(h,l)} \quad \text{(5b)}$$

$$\lambda(h,l) = -J_4 \sum_{\delta,\delta'} a_{r,\delta} a_{s,\delta'} \cos (2\pi [h(\delta+\delta'-1)-l])$$

with Bogoliubov bosonic operators $\tau^\dagger_{\alpha,h,l}$ ($\tau_{\alpha,h,l}$). The neglect of the hardcore property does not concern the large intraladder couplings, but only the small interladder couplings so that the approach is still very accurate [49]. The dispersion $\omega(h,l)$ makes a direct comparison with INS results possible.

To determine the microscopic parameters we fix the value $y = J_1/J_3$ and fit $x = J_2/J_3$, $z = J_4/J_3$, and the energy scale $J_3$ to reproduce the experimental result (eq. (2) in ref. [12])

$$\omega(h,l)^2 = a^2 \cos^2(\pi h) + [\Delta^2 + 4b^2 \sin^2(\pi l)] \sin^2(\pi h) + c^2 \sin^2(2\pi h) \quad \text{(6)}$$

with $a = 4.08(9)$ meV, $\Delta = 1.17(1)$ meV, $b = 0.67(1)$ meV and $c = 2.15(9)$ meV. Thus, we obtain the triples $(x, y, z)$ in table 1. They all essentially imply the same dispersion, see fig. 2. Hence, on the basis of the INS data, one cannot decide which of the triples applies to IPA-CuCl$_3$.

### Table 1: Parameters for IPA-CuCl$_3$ compatible with INS [12].

| $J_3$ [meV] | $x = J_2/J_3$ | $y = J_1/J_3$ | $z = J_4/J_3$ |
|------------|--------------|--------------|-------------|
| 3.743      | 0.133        | -2.0         | -0.076      |
| 3.288      | 0.268        | -1.4         | -0.088      |
| 3.158      | 0.317        | -1.2         | -0.092      |
| 3.038      | 0.369        | -1.0         | -0.096      |
| 2.929      | 0.424        | -0.8         | -0.100      |
| 2.830      | 0.480        | -0.6         | -0.103      |

The quasiparticle decay occurs where the dispersion enters the 2-triplon continuum. It does not prevent to use the CUT for the isolated ladder since the realistic parameters turn out to be such that the triplons do not or hardly decay without the interladder coupling. A quantitative description of the decay is subject of ongoing research.

In complement to the INS we use the temperature dependence of the magnetic susceptibility $\chi(T)$ [26]. Starting from the spin isotropic Hamiltonian (1) the susceptibilities in different spatial directions have to be the same up to scaling proportional to the squares of the Landé $g$-factors. This means that $\chi_A = \chi_B = \chi_C$ equals $g_A^2 = g_B^2 = g_C^2$, where A, B, C indicate the directions normal to the corresponding surfaces of the crystal [26]. Figure 3(a) displays that the three susceptibilities can be scaled to coincide for $g_A = 2.08$, $g_B = 2.06$, and $g_C = 2.25$ within about 3%. This choice of $g$-factors fulfills the experimental constraints [26,27] $g_A, g_B \in [2.06, 2.11]$ and $g_C = 2.25-2.26$ best. We conclude that a spin isotropic Hamiltonian such as (1) provides a very good description, although anisotropies, e.g., Dzyaloshinskii-Moriya terms, can be present with a relative size of a few percent. This agrees with findings from electron paramagnetic resonance [27].
and an asset, we stress that even without the value of \( J_1 \) direction relative to \( \chi_b \) in B and C direction relative to \( \chi_c \) for \( g_B = 2.08, g_B = 2.06, \) and \( g_C = 2.25, \) indicating anisotropies. Lower panel: comparison of \( \chi_B(T) \) for various values \( g_A \) with theoretical results obtained by Dlog-Padé approximated high-temperature series expansions for the \( xyz \) triples from table 1.

Theoretically, we use the high-temperature series expansion for the isolated asymmetric ladder [30] providing series in \( \beta = 1/T \) up to order \( \beta^{n+1} \) with \( n = 10 \) denoted by \( \chi_{1D} \). The 2D series \( \chi_{2D} \) obeys the relation \( \chi_{2D} = \chi_{1D} + J_4 \) in interladder mean-field approximation, i.e., in leading order in \( J_4 \). We use standard Dlog-Padé approximation [50] to deduce the full \( \chi(T) \) from \( \chi_{2D} \) and from the asymptotic behavior \( \chi_{2D}(\beta) \propto \beta^3 \exp(-\Delta \beta) \) for \( 1/\beta \ll \Delta \). The result is plotted in fig. 3 and compared to \( \chi_m \) measured in emu/g and converted according to \( \chi(T) = m_{mol} k_B g \mu_B \omega^{-N_{1D}} \chi_{2D}(T) \). Here \( m_{mol} \) is the molar mass of IPA-CuCl\(_3\), \( k_B \) the Boltzmann constant, \( \mu_B \) the Bohr magneton and \( N_{1D} \) the Avogadro constant.

Figure 3(b) illustrates that theory and experiment agree indeed best for \( g_B = 2.08 \) and the triple of \( y = -0.8 \). As an asset, we stress that even without the value of \( g_B \), the position and the shape of the maximum of \( \chi(T) \) fits best for the triple of \( y = -0.8 \) and one can deduce that the \( g_B \)-factor is around 2.08. As a caveat, we stress the very weak dependence of \( \chi(T) \) on \( y \) in a triple tuned to the INS data. By assuming \( g_B = 2.08 \pm 0.01 \) we estimate the error of our analysis to be \( x = 0.42 \pm 0.06, y = -0.8 \pm 0.2 \) and \( z = -0.100 \pm 0.004 \) implying \( J_1 = -2.3 \pm 0.6 \) meV, \( J_2 = 1.2 \pm 0.2 \) meV, \( J_3 = 2.9 \pm 0.1 \) meV and \( J_4 = -0.292 \) ± 0.001 meV. These values establish the microscopic model for IPA-CuCl\(_3\). We highlight that the ferromagnetic coupling \( J_1 \) does not dominate over the antiferromagnetic coupling \( J_3 \) because \( |y| \ll 1 \), in contrast to the previous purely 1D analysis [26].

The derived microscopic model successfully passes three checks: The BEC is well described, the upper critical field \( H_{c2} \) agrees to experiment and the temperature dependence of the spin gap matches recent data.

First, we follow refs. [38,39,51] to describe the BEC and perform the local transformation

\[
\begin{align}
|s_r\rangle &= u|s_r\rangle + ve^{iQ_0}\langle f|t_{+,r} + g|t_{-,r}\rangle, \quad (7a) \\
|t_{+,r}\rangle &= u\langle f|t_{+,r} + g|t_{-,r}\rangle - ve^{iQ_0}\langle s_r\rangle, \quad (7b) \\
|t_{0,r}\rangle &= |t_{0,r}\rangle, \quad (7c) \\
|t_{-,r}\rangle &= f|t_{-r} - g|t_{+,r}\rangle, \quad (7d)
\end{align}
\]

in real space with \( u = \cos(\theta), \ v = \sin(\theta), \ f = \cos(\varphi) \) and \( g = \sin(\varphi) \), the position \( r = (r, s) \) and the wave vector \( Q_0 = \pi/0 \) of the minimum of the dispersion. The trilong states \( |t_{m}\rangle \) with \( m \in \{-0, +\} \) are given by \( |t_{-}\rangle = 1/\sqrt{2}(|t_{s}⟩ - i|t_{y}\rangle) \), \( |t_{0}\rangle = |t_{z}\rangle \) and \( |t_{+}\rangle = 1/\sqrt{2}(|t_{s}⟩ + i|t_{y}\rangle) \). The trilong states are the same to which the trilong operators in eqs. (2), (3), (4) refer. The tensor product of all singlet states \( |s_r\rangle \) is the vacuum \( |0\rangle \), so that the hardcore trilong creation operator with \( m \in \{-0, +\} \) is defined by \( t_{m,r}^\dagger(0) = |t_{m,r}\rangle \) and the annihilation by \( t_{m,r}^\dagger t_{m,r} : = |0\rangle \) and so on. In this basis the magnetic field is described by the operator \( -h(t_{m,r}^\dagger t_{-} + t_{-}^\dagger t_{m,r}) \). The two independent variables \( \theta \) and \( \varphi \) are varied to minimize the classical ground state energy. This choice also ensures that i) all linear terms in the trilong operators vanish and ii) a massless Goldstone mode appears as it has to be.

Previous work [38,39,51] applied the transformation (7) to the original spin model. This is not possible for IPA-CuCl\(_3\) because the dimers are too strongly coupled. Hence the CUT is mandatory and we apply the real space transformation (7) to \( H_{1D,\text{eff}} + H_{\text{inter,eff}} \) from eqs. (2), (4) keeping the bilinear terms. Fourier transformation and Bogoliubov diagonalization finally provides the lowest lying modes. Their resulting gap energies are displayed in fig. 4. No parameters are adjusted.

Second, the upper critical field \( H_{c2} \) can be determined exactly for the spin model (1a) to be \( H_{c2} = (2J_2 + J_3)/(g\mu_B) \approx 2.9 - 45.8 \) T. After the transformation (7) is applied to the dispersion obtained from CUT we obtain \( H_{c2} \approx 2/g - 45.1 \) T. The very good agreement of these two values strongly supports the approximations made. Additionally, the theoretical values also match the experimental result [31] \( H_{c2} = (43.9 \pm 0.1) \) T(2/g) within 4%. In view of the neglect of anisotropies and magnetoelastic effects, cf. ref. [22], this nice agreement lends independent support to the advocated microscopic model.
Third, the temperature dependence of the gap $\Delta(T)$ supports that the low-lying excitations are hardcore triplons. We apply the mean-field approach in refs. [40,46,52–54] to $H_{1D,\text{eff}} + H_{\text{inter,eff}}$ from eqs. (2), (4). In each nonlocal term ($t_{\text{m,r}} t_{\text{m,r}}^\dagger$ or $t_{\text{m,r}} t_{\text{m,r}}^\dagger$ or $t_{\text{m,r}}^\dagger t_{\text{m,r}}^\dagger$ with $r \neq r'$) all creation operators $t_{\text{m,r}}^\dagger$ are multiplied by the singlet annihilation $s$, and the annihilation operators $t_{\text{m,r}}$ by the singlet creation $s$. Local terms remain unchanged because they do not change the local singlet number. Finally all singlet operators are replaced by the condensate value $s(T) = \langle s \rangle = \langle s \rangle_{\text{cond}}$ with $s \in [0,1]$. In a nutshell, a factor $s^2$ appears in front of each nonlocal term.

This implies a dependence of the dispersion on $s$ and hence on temperature [40,52,54], denoted by $\omega_{\text{disp}}(T)(h,l)$. The self-consistent solution is found from the hardcore condition $1 = \langle s + \sum_{\text{m,r}} t_{\text{m,r}} t_{\text{m,r}}^\dagger \rangle$ leading to $s^2(T) = 1 - 3z/(1 + 3z)$ with

$$z = \int_{-1/2}^{1/2} dh \int_{-1/2}^{1/2} dl \ e^{-\beta\omega_{\text{disp}}(T)(h,l)}.$$  

(8)

Figure 5 compares the result (solid line) of this simple approximation to INS data [32,33]. Up to 15 K the experimental data is matched perfectly. We attribute the discrepancy at higher temperatures to the insufficient treatment of the hardcore constraint by the above approach (for 15 K the condensate fraction $s^2$ is only 0.77). Note that we only apply the mean-field theory to the dispersion obtained from CUT, not to the original spin model as done previously [40,53] because IPA-CuCl$_3$ is not far enough in the dimer limit.

For comparison, we also include $\Delta(T)$ as derived from the nonlinear $\sigma$ model on 1-loop level [55] in fig. 5 (dashed line). It is obtained from

$$C = \int_{-1/2}^{1/2} dh \int_{-1/2}^{1/2} dl \ \frac{\coth(\beta\omega(h,l,T)/2)}{\omega(h,l,T)}$$  

(9)

with $\omega(h,l,T) := \sqrt{\omega^2(h,l) + \Delta^2(T) - \Delta^2(0)}$; the constant $C$ is determined for $T = 0$. Interestingly, this approach describes the experimental data less accurately if the experimental dispersion at $T = 0$ is used for $\omega(h,l)$, cf. ref. [33]. We presume that the hardcore constraint is not accounted for sufficiently well by eq. (9).

In summary, we showed that the available experimental evidence for IPA-CuCl$_3$ is consistent with a quantitative model of weakly coupled asymmetric $S = 1/2$ spin ladders with hardcore triplons as excitations. Such systems are of great current interest because they allow for the study of Bose-Einstein condensation of triplons and of the massless excitations above this condensate [8,9]. Additionally, they represent gapped quantum liquids known to display considerable quasiparticle decay [13–16].

Our high-precision analyses of inelastic neutron scattering data and of the temperature dependence of the magnetic susceptibility is based on advances in continuous unitary transformations [16] and high-temperature series expansions. The established quantitative model paves the way for further quantitative studies, both experimental and theoretical, of the decay of massive quasiparticles and of the condensation of hardcore bosons.

The latter is illustrated by the excellent agreement of the calculated gap energies as function of magnetic field. Additionally, the description of IPA-CuCl$_3$ by dispersive hardcore triplons is strongly supported by the agreement of the temperature dependence of the spin gap.

By this work, a quantitative model for IPA-CuCl$_3$ is established. Concomitantly, we exemplarily showed how CUT results in one dimension at zero temperature and zero magnetic field can be extended to render a quantitative description in two dimensions at finite temperature and finite magnetic field possible. We expect this approach to continue to be fruitful also for other systems.

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