Quasi-universal finite-$T$ scaling in gapped one-dimensional quantum magnets.

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Temperature dependencies of gap energies and magnon lifetimes are measured in the quasi-one-dimensional $S = 1/2$ gapped quantum magnets IP-A–CuCl$_3$ and Sul–Cu$_2$Cl$_4$ using inelastic neutron scattering. The results are compared to those found in literature for $S = 1$ Haldane spin chain materials and to theoretical calculations for the O(3)- and O(N)- quantum non-linear $σ$-models. It is found that when the $T = 0$ energy gap $Δ$ is used as the temperature scale, all experimental and theoretical curves are identical to within system-dependent but temperature-independent scaling factors of the order of unity. This quasi-universality extends over a surprising broad $T$ range, at least up to $κT \sim 1.5Δ$.

One-dimensional (1D) physics is unique in that collisions between counter-propagating particles becomes unavoidable, regardless of their mutual interaction strength. This circumstance has drastic consequences, such as the collapse of the Landau Fermi Liquid in 1D conductors and of long range order in gapless spin chains [1]. In these extreme examples the quasiparticle picture totally breaks down. Not so in gapped quantum spin liquids, such as Haldane spin chains [2] or spin ladders [3, 4]. Here the quasiparticles (magnons) persist in the limit $T \rightarrow 0$, since mutual collisions are rare due to their exponentially small density. Interactions do become important at elevated temperatures thanks to a thermal excitation of a large number of magnons. The result is a reduction of magnon lifetimes and a renormalization of their energies.

In the simplest case the energy gap $Δ$ is large compared to the dispersion bandwidth and magnons are localized. The finite-$T$ effects are then well accounted for by the Random Phase Approximation (RPA) [5, 6]. This approach ignores correlations and breaks down if the quasiparticle are highly mobile, with a velocity $v \gg Δ$. Fortunately, the latter regime is open to investigation using field-theoretical methods. Moreover, as mentioned above, in 1D the details of the interaction potential become irrelevant. For $κT \ll Δ$ a universal theory with a single energy scale $Δ$ can be derived from the large-$S$ mapping of the Heisenberg Hamiltonian onto the O(3) non-linear sigma-model (NLSM) [7, 8]. Until recently, these predictions and the domain of their applicability remained largely untested experimentally. Of the strongly 1D gapped quantum magnets, finite-$T$ data existed only for $S = 1$ Haldane spin chains [9, 10, 11, 12, 13], where $λ = v/Δ \sim 6$ [14]. In the present work we explore the temperature dependence of inelastic neutron spectra in the $S = 1/2$ spin ladder system IP-A–CuCl$_3$ ($λ \sim 2$) [15, 16, 17] and the 4-leg spin tube Sul–Cu$_2$Cl$_4$ ($λ \gtrsim 25$) [18]. We find a striking quasi-universal renormalization of the magnon gaps and lifetimes that persists to surprisingly high temperatures.

As described in detail in [13], the magnetic properties of IP-A–CuCl$_3$ are due to $S = 1/2$ ladders of Cu$^{2+}$ ions that run along the crystallographic $a$ axis of the $P_T$ crystal structure. Exchange interactions along the ladder legs are antiferromagnetic (AF). Pairs of spins on each ladder rung are correlated ferromagnetically. The 1D AF zone center is at $h = 0.5$, where $(h, k, l)$ are components of a wave vector $q$. Due to residual interactions between ladders along the $c$ axis, at low temperatures the spin gap at $h = 0.5$ varies between $Δ = 1.17$ meV for $l = 0$ to $Δ = 1.8$ meV at $l = 0.5$. There is no detectable disper-
The magnon dispersion along the ladder axis is parabolic = study. At low temperatures the gap in Sul–Cu
about 20% of our experimental wave vector resolution,
we discuss here is Sul–Cu
estimate the intrinsic 1D spin ladder gap ∆
is exceptionally 1D. Due to geometric frustration, the 1D
tween adjacent spin tubes are negligible and the system
is 12 times larger than for IPA–CuCl
performed using 3-axis neutron spectrometers. The data
than for Haldane chains.
strong AF leg coupling along the
but comes down to that of 4-leg spin tubes with very
4-meV. In Figs. 3 and
we plot the dimensionless variation of ∆, defined in
as heavy solid curves in Figs. 1a and 2a. The tempera-
ture dependencies of the gap energy ∆(T) obtained from
the fits are plotted in symbols in Figs. 1b and 2b. In
Fig. 3 we plot the temperature-dependent data from [12] in
the two-Lorentzian form, as given by Eqs. 6 and 7
in [10]. Lorentzian line shapes are consistent with the
theoretical results of [8]. The excitation width Γ was
assumed to be q-independent. The dispersion relation was
written as \[ h\omega_0(q) = \Delta^2 + v^2(q\delta - \pi)^2, \]
with \( \delta = a \) for IPA–CuCl3 and \( \delta = c \) for Sul–Cu2Cl4, respectively. The parameters Γ, ∆, and an overall intensity prefactor were
refined to best fit the scans measured at each temperature.
The velocities \( v \) were fixed at their previously determined low-temperature values. Typical fits are plotted
as heavy solid curves in Figs. 1 and 2. The temperature
dependencies of the gap energy ∆(T) obtained from
the fits are plotted in symbols in Figs. 1b and 2b. In
Fig. 3 we plot the dimensionless variation of ∆, defined in
quadrature as \( \delta(T) = \sqrt{\Delta^2(T) - \Delta^2(0)/\Delta_0} \), to emphasize the
low-temperature region. The abscissa axis shows the reduced temperature \( \tau = kT/\Delta_0 \), \( \kappa \) being the Boltz-
man’s constant. The \( \tau \)-dependence of the relative excita-
tion half-width \( \gamma(T) = \Gamma(T)/\Delta_0 \) is shown in Fig. 4 (30).
Note that for IPA–CuCl3 we obtain a perfect overlap be-
tween measurements at the minimum and maximum of the transverse dispersion, validating our expectation that
the temperature dependence is an intrinsic 1D effect.

Central to this study is a comparison of our results for
Sul–Cu2Cl4 and IPA–CuCl3 to those reported in literature
for other 1D quantum spin liquids. Extensive finite-
T data exist for the \( S = 1/2 \) dimer systems Cu(NO3)2
[21] and TiCuCl3 [22]. Unfortunately, in the former compound \( v < \Delta \), while the latter is by no measure one-
dimensional. Among the known materials that do fit our
requirements, neutron data exist only for \( S = 1 \) Haldane
spin chains. CsNiCl3 is one of the oldest known prototypes [23, 24]. Inter-chain interactions in this compound
are significant, and it actually orders in 3D at \( T = 4.8 \) K. At the 3D zone-center the gap softens at the transition
point [10], but one can hope to retrieve 1D behavior at certain special wave vectors [12]. The intrinsic 1D gap
energy in CsNiCl3 is \( \Delta(0) \sim 0.9 \) meV. In Figs. 3 and
4 we plot the temperature-dependent data from [12] in
star symbols. The organo-metallic complexes NENP [25]
and NINAZ [11] are excellent 1D systems, but they are
affected by the rather strong magnetic anisotropy. This
anisotropy splits the triplet of Haldane gap modes into

\[
\begin{align*}
\Delta_0 &= 1.5 \text{ meV} \\
\lambda &= v/\Delta_0 = 1.9 \\
\lambda &= 25 \\
\lambda &= 12 \text{ times larger than for IPA–CuCl3 and 4 times larger than for Haldane chains.} \\
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\lambda &= 12 \text{ times larger than for IPA–CuCl3 and 4 times larger than for Haldane chains.} \\
\end{align*}
\]
dependence of the gap energy in the Fig. 3 shows a one-loop calculation of the temperature described above to field-theoretical results. The solid line [19]. For this model thermal broadening of excitations on IPA–CuCl$_3$NENP [25], and λ⊥ respectively. The most recent and extensive study [13] is on NENP, NINAZ, and Y$_2$BaNiO$_5$ data are from Refs. [12, 3, 11], and 13, respectively. Calculations for the O(3) and O(N) non-linear σ-models are from Refs. [19] and [20], correspondingly.

A low energy transverse-polarized doublet and a longitudinal higher-energy singlet [26], with gaps Δ⊥ and Δ∥, respectively. As argued in [19], in this case one can still make a meaningful comparison with Heisenberg systems by focusing on the two lower modes. For NENP and NINAZ Δ⊥ = 1.1 meV and Δ∥ = 3.6 meV, respectively. In Figs. 3 and 4 we plot δ(T) = \sqrt{Δ^2(T) − Δ^2(0)}/Δ(0) and γ(T) = Γ(Δ)/Δ(0) vs. τ = T/Δ(0), with up and down triangles for NENP [4] and NINAZ [11], respectively. The most recent and extensive study [13] is on Y$_2$BaNiO$_5$ that is known to be an excellent 1D Haldane gap system with Δ = 8.6 meV and only weak anisotropy [27]. In Figs. 3 and 4 the corresponding data from [13] are plotted in square symbols. As mentioned above, for S = 1 Haldane spin chains numerical calculations predict λ = 6.2 [13], but it is also useful to quote the experimental ratio λ ≡ ν/Δ = 6.4 for CsNiCl$_3$ [24], λ = 6.1 for NENP [25], and λ = 8.1 for Y$_2$BaNiO$_5$ [27].

It is instructive to compare the experimental data on IPA–CuCl$_3$, Sul–Cu$_2$Cl$_4$ and the other materials described above to field-theoretical results. The solid line in Fig. 3 shows a one-loop calculation of the temperature dependence of the gap energy in the O(3)-NLSM from [19]. For this model thermal broadening of excitations was calculated in [8], and is plotted in a dash-dot line in Fig. 3. More results are available for the large-N limit of the NLSM that is more easily tractable. O(N)-NLSM calculations for Δ(T) were reported in [20] and are plotted in a dashed line in Fig. 3.

The thermal renormalization of gap energy remains within a factor of two for all the diverse materials and models considered. From the theoretical viewpoint, the increase is driven by a repulsion between thermally excited quasiparticles [7, 8, 20]. It is proportional to the correlation length λ and the quasiparticle density ρ. At low temperatures the latter universally scales as ρ \propto √τ/λ exp(-1/τ) for all 1D systems with a dynamically generated gap. As a result, for κT ≪ Δ one can expect all measured δ(τ) dependencies, as well as the ones calculated for the O(3)- and O(N)-NLSM, to be the same, to within a system-dependent but temperature-independent scaling factor α. The latter will reflect the details of quasiparticle interactions in each particular model. Experimentally, this is indeed the case in the entire studied temperature range. The inset in Fig. 3 shows such a scaling plot, where we have arbitrarily selected α = 1 for the O(N)-NLSM, α = 1.3 for the spin ladders in IPA–CuCl$_3$, α = 1.35 for Sul–Cu$_2$Cl$_4$, and α = 2.35 for all the Haldane spin chains. The data on CsNiCl$_3$ were excluded from the scaling, as they are inconsistent with those on other S = 1 chains, due to the strong 3D interactions in that material [10].

A similar quasi-universal scaling can be also expected for the excitation widths. The latter is given by the inverse average time between collisions among thermally excited quasiparticles and is thus proportional to ρ and
thermal average of quasiparticle velocity $\nabla \propto v \sqrt{T/\Delta}$ [8]. The experimental curves in Fig. 4 are indeed very close for all systems under consideration, and a near-perfect collapse (inset) is obtained with scaling factors $\beta = 1$ for the O(N)-NLSM, $\beta = 1.2$ for the spin ladders in IPA–CuCl$_3$, $\beta = 0.8$ for Sul–Cu$_2$Cl$_4$, and $\beta = 1.6$ for Haldane chains.

Deviations from unity of the factors $\alpha$ and $\beta$ reflect the inadequacy of the NLSM mapping. Indeed, it is rigorous only in the limit $S \gg 1$. As evident from recent numerical studies [23], that model has only limited quantitative applicability to $S = 1/2$ and $S = 1$ chains. An alternative description of the $S = 1/2$ ladder contains quasiparticle interactions as a free parameter [20], and may be more versatile. Of course, any quasi-universal behavior with the energy scale set by $\Delta$ will have a solid theoretical justification only in the limit $\kappa T \ll \Delta$. A different universality is expected for $\nu \gg \kappa T \gg \Delta$, where the field theoretical description is still justified, but the spin gap can be considered negligible. Here the only remaining energy scale is the temperature itself and $\chi''(\pi, \omega)$ is a function of $\omega/T$. Most of our data actually lie in the crossover region $\kappa T \sim \Delta$, while access to the limit $\kappa T \ll \Delta$ is hindered by the limited experimental resolution. The two central experimental findings are thus the quasi-universal $T/\Delta$ scaling in vastly diverse models and real small-$S$ materials, and its persistence well beyond the low-temperature limit, up to at least $\kappa T \sim 1.5 \Delta$.

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