Extended Universal Finite-T Renormalization of Excitations in a Class of One-Dimensional Quantum Magnets

A. Zheludev,[1,*] V. O. Garlea,[1] L.-P. Regnault,[2] H. Manaka,[3] A. Tsvelik,[4] and J.-H. Chung[5,†]

[1]*Neutron Scattering Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*
[2]*CEA-Grenoble, DRFMC-SPSMS-MDN, 17 rue des Martyrs, 38054 Grenoble Cedex 9, France*
[3]*Graduate School of Science and Engineering, Kagoshima University, Korimoto, Kagoshima 890-0065, Japan*
[4]*Brookhaven National Laboratory, Upton, New York 11973, USA*
[5]*NCNR, National Institute of Standards and Technology, Gaithersburg, Maryland 20899 and University of Maryland, College Park, Maryland, 20742, USA*

(Received 26 November 2007; revised manuscript received 13 February 2008; published 16 April 2008)

Temperature dependencies of gap energies and magnon lifetimes are measured in the quasi-one-dimensional $S = 1/2$ gapped quantum magnets $(CH_3)_2CHNH_3CuCL_3$ (IPA-$CuCl_3$, where IPA denotes isopropyl ammonium) and $Cu_2Cl_4 \cdot D_8C_4SO_2$ (Sul-Cu_2Cl_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 nonlinear 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 $\kappa T \sim 1.5\Delta$.

DOI: 10.1103/PhysRevLett.100.157204 PACS numbers: 75.10.Pq, 75.40.Gb

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 $v \gg \Delta$. 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 $\kappa T \ll \Delta$, a universal theory with a single energy scale Δ can be derived from the large-S mapping of the Heisenberg Hamiltonian onto the $O(3)$ nonlinear 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 [9–13], where $\lambda = v/\Delta \sim 6$ [14]. In the present Letter, we explore the temperature dependence of inelastic neutron spectra in the $S = 1/2$ spin ladder system $(CH_3)_2CHNH_3CuCL_3$ (IPA-$CuCl_3$, where IPA denotes isopropyl ammonium) ($\lambda \sim 2$) [15–17] and the 4-leg spin tube $Cu_2Cl_4 \cdot D_8C_4SO_2$ (Sul-Cu_2Cl_4) ($\lambda \gtrsim 25$) [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 [15], the magnetic properties of IPA-$CuCl_3$ are due to $S = 1/2$ ladders of Cu^{2+} ions that run along the crystallographic a axis of the $P\bar{1}$ crystal structure. Exchange interactions along the ladder legs are antiferromagnetic (AFM). Pairs of spins on each ladder rung are correlated ferromagnetically. The 1D AFM zone center is at $h = 0.5$, where (h, k, l) are components of a wave vector $\mathbf{q}$. Because of residual interactions between ladders along the c axis, at low temperatures, the spin gap at $h = 0.5$ varies between $\Delta = 1.17$ meV for $l = 0$ to $\Delta = 1.8$ meV at $l = 0.5$. There is no detectable dispersion of magnetic excitations along the b axis. Treating the weak interladder coupling at the RPA level, one can estimate the intrinsic 1D spin ladder gap $\Delta_0 = 1.5$ meV. The magnon dispersion along the ladder axis is parabolic at the 1D AFM zone center and characterized by a velocity $v = 2.9$ meV, as defined by Eq. 2 in [17], so that $\lambda = v/\Delta_0 = 1.9$. The other Cu^{2+} based material that we discuss here is Sul-Cu_2Cl_4 [18]. The corresponding topology of its magnetic interactions is rather complex, but comes down to that of 4-leg spin tubes with very strong AFM leg coupling along the c axis. Interactions between adjacent spin tubes are negligible, and the system is exceptionally 1D. Because of geometric frustration, the 1D AFM zone center is located at slightly incommensurate position $l = 0.5 - \zeta$, where $\zeta = 0.022(2)$. ζ amounts to only about 20% of our

experimental wave vector resolution, and can be safely ignored in the context of the present study. At low temperatures, the gap in Sul-Cu_2Cl_4 is $\Delta_0 = 0.55$ meV. Dispersion along the c^* direction is very steep, with $v = 14$ meV. Thus, for Sul-Cu_2Cl_4, $\lambda = 25$ is 12 times larger than for IPA-$CuCl_3$ and 4 times larger than for Haldane chains.

Temperature-dependent inelastic measurements were performed using 3-axis neutron spectrometers. The data on IPA-$CuCl_3$ at momentum transfer $\mathbf{q} = (0.5, 0, 0)$ were collected on the IN22 instrument at ILL using 5 meV fixed-final neutron energy, a pyrolitic graphite (PG) monochromator, a horizontally focused PG analyzer, and a Be filter after the sample. The background was measured independently at each temperature away from the 1D AFM zone-center, at $\mathbf{q} = (0.3, 0, 0)$ and $\mathbf{q} = (0.7, 0, 0)$. The sample consisted of 20 coaligned fully deuterated single crystals of total mass 3 g. Sample environment was a standard He-flow cryostat. A similar configuration on the NG-5 SPINS instrument at NIST was used with 3.7 meV fixed final energy neutrons to collect data around (0.5, 0, 0.5) in IPA-$CuCl_3$ and at (0, 0, 0.5) in Sul-Cu_2Cl_4. For the latter material, we utilized a $\sim 2g$ assembly of 15 coaligned deuterated single crystals and a ^{4}He-^{3}He dilution refrigerator.

Typical background-subtracted energy scans collected at the 1D AFM zone-centers in IPA-$CuCl_3$ and Sul-Cu_2Cl_4 at different temperatures are shown in Figs. 1(a) and 2(a), respectively. Series of such scans are assembled into false-color plots in Figs. 1(b) and 2(b). Here, the measured intensity at each temperature was normalized by its peak value. The observed gap modes broaden with temperature, and the gap energies increase. The data were fit to a model cross section function that was numerically convoluted with the resolution function of the spectrometer. The dynamic structure factor was written 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 $\mathbf{q}$-independent. The dispersion relation was written as $[\hbar\omega_0(\mathbf{q})]^2 = \Delta^2 + v^2(\mathbf{q}.\mathbf{d} - \pi)^2$, with $\mathbf{d} = \mathbf{a}$ for IPA-$CuCl_3$ and $\mathbf{d} = \mathbf{c}$ for Sul-Cu_2Cl_4, respectively. The parameters Γ, Δ, 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(a) and 2(a). The temperature dependencies of the gap energy $\Delta(T)$ obtained from the fits are plotted in symbols in Figs. 1(b) and 2(b). In Fig. 3, we plot the dimensionless variation of Δ, 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 $\tau = \kappa T/\Delta_0$, κ being the Boltzman's constant. The τ-dependence of the relative excitation half-width $\gamma(T) = \Gamma(T)/\Delta_0$ is shown in Fig. 4 [19]. Note that for IPA-$CuCl_3$, we obtain a perfect overlap between 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-Cu_2Cl_4 and IPA-$CuCl_3$ to those reported in literature for other 1D quantum spin liquids. Extensive finite-T data exist for the $S = 1/2$ dimer systems $Cu(NO_3)_2$ [20] and $TlCuCl_3$ [21]. Unfortunately, in the former compound $v \ll \Delta$, 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 chain systems. $CsNiCl_3$ is one of the oldest known prototypes [22,23]. Interchain interactions in this compound are significant, and it actually orders in 3D at $T = 4.8$ K. At the

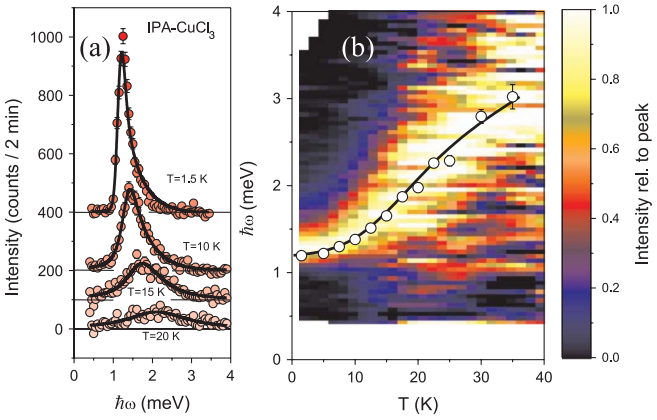

FIG. 1 (color). (a) Typical background-subtracted constant-q scans collected at the 1D zone-center (0.5, 0, 0) in the spin ladder compound IPA-$CuCl_3$ ($v/\Delta = 1.9$) at different temperatures (symbols). Solid lines are fits to the data, as described in the text. (b) False-color representation of inelastic intensity measured at the 1D AFM zone center in IPA-$CuCl_3$ plotted as a function of temperature and energy transfer. Symbols are the measured temperature dependence of the gap energy. The solid line is a guide for the eye.

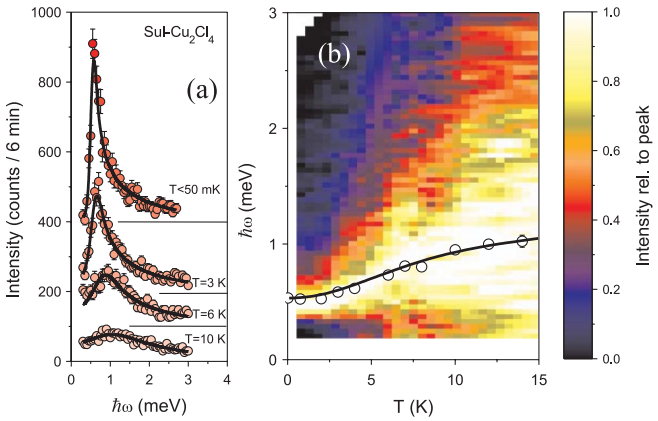

FIG. 2 (color). Same as Fig. 1, but for the (0, 0, 0.5) 1D 4-leg spin-tube material Sul-Cu_2Cl_4 ($v/\Delta > 20$).

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 $CsNiCl_3$ 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 $Ni(C_2H_8N_2)_2NO_2ClO_4$ (NENP) [24] and $Ni(C_3H_{10}N_2)_2N_3(ClO_4)$ (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 a low energy transverse-polarized doublet and a longitudinal higher-energy singlet [25], with gaps $\Delta_\perp$ and $\Delta_\parallel$, respectively. As argued in [26], in this case, one can still make a meaningful comparison with Heisenberg systems by focusing on the two lower modes. For NENP and NINAZ, $\Delta_\perp = 1.1$ meV and $\Delta_\perp = 3.6$ meV, respectively. In Figs. 3 and 4, we plot $\delta(T) = \sqrt{\Delta_\perp^2(T) - \Delta_\perp^2(0)}/\Delta_\perp(0)$ and $\gamma(T) = \Gamma_\perp(T)/\Delta_\perp(0)$ vs $\tau = T/\Delta_\perp(0)$, with up and down triangles for NENP [9] and NINAZ [11], respectively. The most recent and extensive study [13] is on Y_2BaNiO_5 that is known to be an excellent 1D Haldane gap system with $\Delta = 8.6$ 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 $\lambda = 6.2$ [14], but it is also useful to quote the experimental ratio $\lambda \equiv v/\Delta = 6.4$ for $CsNiCl_3$ [23], $\lambda = 6.1$ for NENP [24], and $\lambda = 8.1$ for Y_2BaNiO_5 [27].

It is instructive to compare the experimental data on IPA-$CuCl_3$, Sul-Cu_2Cl_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 [26]. For this model, thermal broadening of excitations was calculated in [8] and is plotted in a dash-dot line in Fig. 4. More results are available for the large-N limit of the NLSM that is more easily tractable. $O(N)$NLSM calculations for $\Delta(T)$ were reported in [28] and are plotted in a dashed line in Fig. 3.

The thermal renormalization of gap energy remains within a factor of 2 for all the diverse materials and models considered. From the theoretical viewpoint, the increase is driven by a repulsion between thermally excited quasiparticles [7,8,28]. It is proportional to the correlation length λ and the quasiparticle density ρ. At low temperatures, the latter *universally* scales as $\rho \propto \sqrt{\tau}/\lambda \exp(-1/\tau)$ for all 1D systems with a dynamically generated gap. As a result, for $\kappa T \ll \Delta$ one can expect all measured $\delta(\tau)$ 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 $\alpha = 1$ for the $O(N)$NLSM, $\alpha = 1.3$ for the spin ladders in IPA-$CuCl_3$, $\alpha = 1.35$ for Sul-Cu_2Cl_4, and

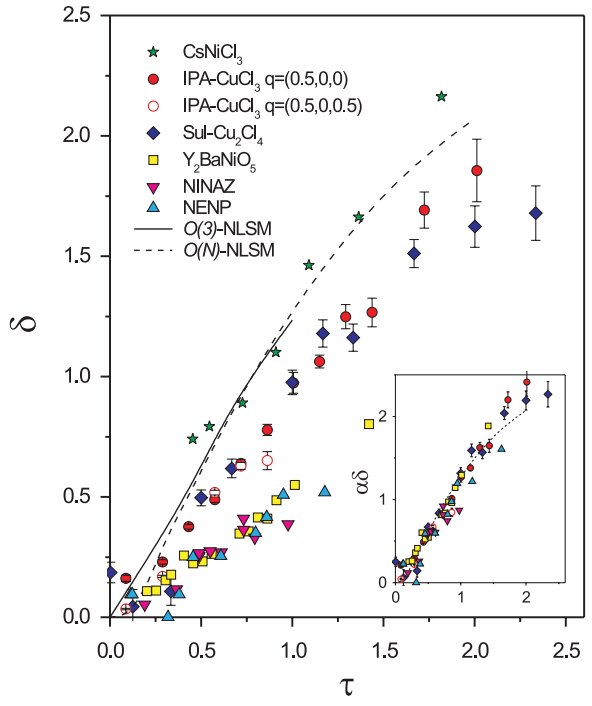

FIG. 3 (color). Reduced gap energy δ plotted against reduced temperature τ for diverse quasi 1D quantum antiferromagnetic materials (symbols) and analytical models (lines). Inset: same data scaled to show universal behavior. $CsNiCl_3$, $Ni(C_2H_8N_2)_2NO_2ClO_4$ (NENP), $Ni(C_3H_{10}N_2)_2N_3(ClO_4)$ (NINAZ), and Y_2BaNiO_5 data are from Refs. [9,11–13], respectively. Calculations for the $O(3)$ and $O(N)$ nonlinear σ-models are from Refs. [26,28], correspondingly.

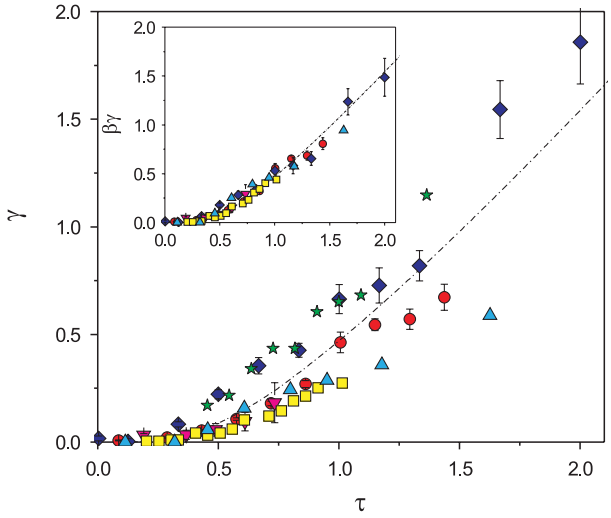

FIG. 4 (color). Reduced energy width γ of the gap excitations plotted against reduced temperature τ for diverse quasi 1D quantum antiferromagnetic materials (symbols, as in Fig. 3) and the $O(3)$ nonlinear σ-model (dash-dot line, Ref. [8]).

$\alpha = 2.35$ 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 $\bar{v} \propto v\sqrt{T/\Delta}$ [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_2Cl_4, and $\beta = 1.6$ for Haldane chains.

Deviations from unity of the factors α and β reflect the inadequacy of the NLSM mapping. Indeed, it is rigorous only in the limit $S \gg 1$. As evident from recent numerical studies [29], 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 [30], and may be more versatile. Of course, any quasi-universal behavior with the energy scale set by Δ will have a solid theoretical justification only in the limit $\kappa T \ll \Delta$. A different universality is expected for $v \gg \kappa T \gg \Delta$, 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 ω/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/Δ scaling in vastly diverse models and real small-S materials, and its persistence well beyond the low-temperature limit, up to at least $\kappa T \sim 1.5\Delta$.

One of the authors (A. Z.) would like to thank F. Essler, I. Zanliznyak, and Ch. Ruegg for insightful and educational discussions. Research at ORNL and BNL was funded by the United States Department of Energy, Office of Basic Energy Sciences- Materials Science, under Contract Nos. DE-AC05-00OR22725 and DE-AC02-98CH10886, respectively. The work at NCNR was supported by the National Science Foundation under Agreement Nos. DMR-9986442, -0086210, and -0454672.

*zheludevai@ornl.gov

†Present address: Department of Physics, Korea University Anam-dong 5, Seongbook-gu Seoul 126-713, Rep. of Korea

[1] A. Tsvelik, *Quantum Field Theory in Condensed Matter Physics* (Cambridge University Press, Cambridge, 2007).
[2] F. D. M. Haldane, Phys. Lett. A **93**, 464 (1983); Phys. Rev. Lett. **50**, 1153 (1983).
[3] T. M. Rice, S. Gopalan, and M. Sigrist, Europhys. Lett. **23**, 445 (1993).
[4] T. Barnes, E. Dagotto, J. Riera, and E. S. Swanson, Phys. Rev. B **47**, 3196 (1993).
[5] B. Leunberger, A. Stebler, H. U. Gudel, A. Furrer, R. Feile, and J. K. Kjems, Phys. Rev. B **30**, 6300 (1984).
[6] Y. Sasago, K. Uchinokura, A. Zheludev, and G. Shirane, Phys. Rev. B **55**, 8357 (1997).
[7] S. Sachdev and K. Damle, Phys. Rev. Lett. **78**, 943 (1997).
[8] C. Damle and S. Sachdev, Phys. Rev. B **57**, 8307 (1998).
[9] J. Renard, M. Verdaguer, L. Regnault, W. Erkelens, J. Rossat-Mignod, J. Ribas, W. Stirling, and C. Vettier, J. Appl. Phys. **63**, 3538 (1988).
[10] I. A. Zaliznyak, L.-P. Regnault, and D. Petitgrand, Phys. Rev. B **50**, 15824 (1994).
[11] A. Zheludev, S. E. Nagler, S. M. Shapiro, L. K. Chou, D. R. Talham, and M. W. Meisel, Phys. Rev. B **53**, 15004 (1996).
[12] M. Kenzelmann, R. A. Cowley, W. J. Buyers, R. Coldea, M. Enderle, and D. F. McMorrow, Phys. Rev. B **66**, 174412 (2002).
[13] G. Xu, C. Broholm, Y.-A. Soh, G. Aeppli, J. F. DiTusa, M. K. Y. Chen, C. D. Frost, T. Ito, K. Oka, and H. Takagi, Science **317**, 1049 (2007).
[14] S. White and D. Huse, Phys. Rev. B **48**, 3844 (1993).
[15] T. Masuda, A. Zheludev, H. Manaka, L.-P. Regnault, J.-H. Chung, and Y. Qiu, Phys. Rev. Lett. **96**, 047210 (2006).
[16] V. O. Garlea, A. Zheludev, T. Masuda, H. Manaka, L.-P. Regnault, E. Ressouche, B. Grenier, J.-H. Chung, Y. Qiu, K. Habicht *et al.*, Phys. Rev. Lett. **98**, 167202 (2007).
[17] A. Zheludev, V. O. Garlea, T. Masuda, H. Manaka, L.-P. Regnault, E. Ressouche, B. Grenier, J.-H. Chung, Y. Qiu, K. Habicht *et al.*, Phys. Rev. B **76**, 054450 (2007).
[18] V. O. Garlea, A. Zheludev, L.-P. Regnault, J.-H. Chung, Y. Qiu, and M. Boehm, Phys. Rev. Lett. **100**, 037206 (2008).
[19] For $T > 25$ K, the quality of the data for IPA-$CuCl_3$ was not sufficient to simultaneously fit Δ and Γ as independent parameters. In that regime only Δ was refines, while Γ was fixed to 1.2 meV.
[20] G. Xu, C. Broholm, D. H. Reich, and M. A. Adams, Phys. Rev. Lett. **84**, 4465 (2000).
[21] C. Ruegg, B. Normand, M. Matsumoto, C. Niedermayer, A. Furrer, K. W. Krämer, H.-U. Güdel, P. Bourges, Y. Sidis, and H. Mutka, Phys. Rev. Lett. **95**, 267201 (2005).
[22] W. J. L. Buyers, R. Morra, R. L. Armstrong, M. J. Hogan, P. Gerlach, and K. Hirakawa, Phys. Rev. Lett. **56**, 371 (1986).
[23] R. M. Morra, W. J. L. Buyers, R. L. Armstrong, and K. Hirakawa, Phys. Rev. B **38**, 543 (1988).
[24] L. P. Regnault, I. Zaliznyak, J. P. Renard, and C. Vettier, Phys. Rev. B **50**, 9174 (1994).
[25] O. Golinelli, T. Jolicoeur, and R. Lacaze, Phys. Rev. B **45**, 9798 (1992).
[26] D. Senechal, Phys. Rev. B **47**, 8353 (1993).
[27] G. Xu, J. F. DiTusa, T. Ito, H. Takagi, C. L. Broholm, and G. Aeppli, Phys. Rev. B **54**, R6827 (1996).
[28] T. Jolicoeur and O. Golinelli, Phys. Rev. B **50**, 9265 (1994).
[29] S. R. White and I. Affleck, arXiv:0712.3785.
[30] D. Shelton, A. A. Nersesyan, and A. M. Tsvelik, Phys. Rev. B **53**, 8521 (1996).