Self-consistent theory for the paramagnetic properties of $TlCuCl_3$ to first order in the high-density $1/z$ expansion

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(Received 15 October 2010; revised manuscript received 6 December 2010; published 25 February 2011)

The Cu spins in $TlCuCl_3$ constitute a well-defined dimer system, where the interaction between the dimers is just at the threshold for inducing magnetic order. The leading order effects of the fluctuations on the paramagnetic properties are analyzed in terms of the systematic high-density $1/z$ expansion. The predictions agree with those derived from the self-consistent RPA theory published in Phys. Rev. B **80**, 224419 (2009). Linewidth effects were neglected in the previous theory, whereas the present one accounts for both the energies and the linewidths of the singlet-triplet excitations. These results are found to be in good agreement with experimental observations at zero field. Experimentally, the renormalization effects are enhanced at fields close to the critical one. This is in accord with the predictions, but both theories overestimate the enhancement.

DOI: 10.1103/PhysRevB.83.064420 PACS number(s): 75.10.−b, 75.30.−m, 67.85.Jk

I. INTRODUCTION

The $S = 1/2$ spins of the Cu^{2+} ions in $TlCuCl_3$ are strongly coupled pairwise. The coupling is antiferromagnetic and the ground state of the isolated dimers is the $S = 0$ ground state with a gap $\Delta \approx 5.2$–5.7 meV up to the $S = 1$ triplet. The dimer system is close to a quantum critical point, and a zero-temperature phase transition may be achieved either by the application of a modest magnetic field of 54 kOe[1] or hydrostatic pressure of 1.1 kbar.[2,3] These circumstances make the present dimer system unique, as it clearly exhibits the importance of quantum and thermal fluctuations (see, for instance, the review by Giamarchi *et al.*[4]).

The system has been analyzed several times in the past using different theoretical methods. Matsumoto *et al.*[5] applied the random-phase approximation (RPA) to calculate all zero-temperature properties of $TlCuCl_3$. Nikuni *et al.*[6] utilized the parallel between the present spin system and a system of massive bosons for mapping the field-induced phase transition on the transition to a Bose-Einstein condensate (BEC).[7] Later on, a more realistic BEC description of the system was presented by Misguich and Oshikawa,[8] and Sirker *et al.*[9] used a bond-operator approach to map the spin system onto a model of interacting bosons with an infinite on-site repulsion between the triplet excitations. Although the BEC analog offers a way for determining the influences of the fluctuations, these theories are not entirely satisfactory. The BEC mapping does not specify the v_0 interaction between the bosons. The assumption made by Nikuni and coworkers, that only the lowest of the local triplet levels is important for determining the condensate, is an oversimplification. The *ad hoc* assumption, that the prevention of double occupancy of a local level is accounted for by a simple repulsive potential, might be too imprecise.

In a previous paper, Ref. 10, the influences of the spin fluctuations in $TlCuCl_3$ were analyzed in the neighborhood of the field-induced phase transition in a direct fashion with no use of the BEC analogy. This self-consistent RPA (ScRPA) theory handled the problem of double occupancy of the local dimer levels by including the corrections appearing in the hierarchy of decoupled equations of motion in the next order beyond the RPA. The theory did not rely on any adjustable parameters, as all results were expressed in terms of the exchange parameters in the spin Hamiltonian. These parameters were then derived from the zero-field and zero-temperature excitation spectrum measured in inelastic neutron-scattering experiments.[11,12]

In the present paper the effects due to fluctuations are included by utilizing the high-density $1/z$ expansion[13] within the effective-medium approach.[14,15] The expansion parameter is defined by the property that every $\vec{q}$ summation over reciprocal space effectively supplies a factor $1/z$, where z is the coordination number of interacting neighbors.[13] To zero order, the fluctuations are neglected and the theory is identical with the mean-field RPA theory. To first order the theory includes the effects of the fluctuations in the surroundings of each site. These single-site fluctuations may be accounted for in a self-consistent manner, since the fluctuating surroundings, to first order in $1/z$, constitute an "effective medium" that is common for all sites.[14,15] This $1/z$-expansion theory has previously been applied to the singlet-ground state systems Pr and HoF_3 and to the Ising ferromagnet $LiHoF_4$.[14–17] Here we shall consider the case of paramagnetic $TlCuCl_3$ in a magnetic field at ambient pressure.

II. THEORY

The effective spin Hamiltonian for paramagnetic $TlCuCl_3$, in the presence of a magnetic field, is established in Ref. 10 and is

$$\begin{aligned}\mathcal{H} = &\sum_i \left[\Delta_1 a^i_{11} + \Delta_2 a^i_{22} + \Delta_3 a^i_{33}\right] \\ &- \frac{1}{2}\sum_{ij} J(ij)\left[\left(a^i_{02} + a^i_{20}\right)\left(a^j_{02} + a^j_{20}\right)\right. \\ &\left. + \left(a^i_{01} - a^i_{30}\right)\left(a^j_{10} - a^j_{03}\right) + \left(a^i_{10} - a^i_{03}\right)\left(a^j_{01} - a^j_{30}\right)\right] \quad (1)\end{aligned}$$

in terms of the standard basis operators $a^i_{\nu\mu} = (|\nu\rangle\langle\mu|)_i$. $|\nu\rangle$, $\nu = 0, \ldots, 3$, denote the mean-field eigenstates of the ith dimer, and the corresponding eigenenergies are

$$\Delta_1 = \Delta - h, \quad \Delta_2 = \Delta, \quad \Delta_3 = \Delta + h. \quad (2)$$

Δ is the zero-field splitting between the singlet and the triplet states of a noninteracting dimer, and $h = g\mu_B H$.

The two-site Green's functions are defined as the τ-ordered ensemble averages

$$G_{\xi\eta,\xi'\eta'}(ij,\tau) = -\left\langle T_\tau a^i_{\xi\eta}(\tau) a^j_{\xi'\eta'}(0)\right\rangle. \tag{3}$$

The Green's functions involved in the effective coupling matrix [Eq. (2.13) in Ref. 16] are

$$\begin{aligned} \mathcal{G}_2 &= G_{02,20} + G_{02,02} + G_{20,02} + G_{20,20}\,, \\ \mathcal{G}_{31} &= G_{03,30} + G_{10,01} - G_{03,01} - G_{10,30}\,, \\ \mathcal{G}_{13} &= G_{30,03} + G_{01,10} - G_{30,10} - G_{01,03}. \end{aligned} \tag{4}$$

Performing a Fourier transformation in τ and space, all the Green's functions attain the common argument $i\omega_n$, where ω_n is the discrete Matsubara frequency $2\pi n/\beta$ (where $\beta = 1/k_BT$), and it is straightforward to show that $\mathcal{G}_{13}(\vec{q},i\omega_n) = \mathcal{G}_{31}(\vec{q},-i\omega_n)$.

To first order in $1/z$, the self-consistent, effective-medium result for the final Green's functions is[16]

$$\mathcal{G}_p(\vec{q},i\omega_n) = \frac{\mathcal{G}_p(i\omega_n)}{1 + [J(\vec{q}) - K_p(i\omega_n)]\mathcal{G}_p(i\omega_n)}\,, \tag{5}$$

where the index $p = 2$, 13, or 31. The effective-medium coupling parameters are $K_2(i\omega_n) = K_2(-i\omega_n)$ and $K_{31}(i\omega_n) = K_{13}(-i\omega_n)$, and they are determined by the condition that the single-site Green's functions, per definition, are

$$\mathcal{G}_p(i\omega_n) = \frac{1}{N}\sum_{\vec{q}} \mathcal{G}_p(\vec{q},i\omega_n) \tag{6}$$

or

$$K_p(i\omega_n) = \sum_{\vec{q}} J(\vec{q})\,\mathcal{G}_p(\vec{q},i\omega_n) \Bigg/ \sum_{\vec{q}} \mathcal{G}_p(\vec{q},i\omega_n). \tag{7}$$

Hence the final Green's functions in Eq. (5) are determined by the interaction $J(\vec{q})$, when knowing the single-site ones, $\mathcal{G}_p(i\omega_n)$. The terms in the cumulant expansion of the single-site Green's functions [Eq. (2.15) in Ref. 16] may be calculated by utilizing the generalized Wick's theorem derived by Care and Tucker,[18] which involves the propagators

$$g_{\xi\eta}(i\omega_n) = \frac{1}{(\Delta_\eta - \Delta_\xi) - i\omega_n} \tag{8}$$

($\Delta_0 = 0$). Assuming a "Dyson-like" behavior of the cumulant expansion,[16] the propagators are found to be renormalized according to

$$\widetilde{g}_{\xi\eta}(i\omega_n) = \frac{1}{(\widetilde{\Delta}_\eta - \widetilde{\Delta}_\xi) - i\omega_n}, \quad \widetilde{\Delta}_\xi = \Delta_\xi + \delta_\xi(i\omega_n) \tag{9}$$

[$\delta_0(i\omega_n) = 0$]. Introducing the mean-field population factors

$$n_\xi = e^{-\beta\Delta_\xi} \Bigg/ \sum_{\eta=0}^{3} e^{-\beta\Delta_\eta} \quad \text{and} \quad n_{\xi\eta} = n_\xi - n_\eta, \tag{10}$$

the longitudinal component of the single-site Green's functions is determined as

$$\mathcal{G}_2(i\omega_n) = \frac{\mathcal{G}^0_2(i\omega_n)}{1 + [K_2(i\omega_n) - \gamma_2(i\omega_n)]\mathcal{G}^0_2(i\omega_n)}\,, \tag{11}$$

where

$$\mathcal{G}^0_2(i\omega_n) = -\frac{n_{02}}{1+\alpha_2}[\widetilde{g}_{02}(i\omega_n) - \widetilde{g}_{20}(i\omega_n)]. \tag{12}$$

Similarly, the two transverse components, $\mathcal{G}_{31}(i\omega_n)$ and $\mathcal{G}_{13}(i\omega_n) = \mathcal{G}_{31}(-i\omega_n)$, are determined from

$$\mathcal{G}_{31}(i\omega_n) = \frac{\mathcal{G}^0_{31}(i\omega_n)}{1 + [K_{31}(i\omega_n) - \gamma_{31}(i\omega_n)]\mathcal{G}^0_{31}(i\omega_n)}\,, \tag{13}$$

where

$$\mathcal{G}^0_{31}(i\omega_n) = -\frac{n_{03}}{1+\alpha_3}\widetilde{g}_{03}(i\omega_n) + \frac{n_{01}}{1+\alpha_1}\widetilde{g}_{10}(i\omega_n). \tag{14}$$

The different parameters introduced above are determined in terms of a number of frequency sums. The longitudinal ones are

$$\lambda^L_s = \frac{1}{2\beta}\sum_n K_2(i\omega_n)([n_{02}g_{02}(i\omega_n)]^s + [-n_{02}g_{20}(i\omega_n)]^s), \tag{15}$$

where s is either 1 or 2. The transverse sums are

$$\begin{aligned} \lambda^{T1}_s &= \frac{1}{\beta}\sum_n K_{31}(i\omega_n)[-n_{01}g_{10}(i\omega_n)]^s, \\ \lambda^{T3}_s &= \frac{1}{\beta}\sum_n K_{31}(i\omega_n)[n_{03}g_{03}(i\omega_n)]^s. \end{aligned} \tag{16}$$

The renormalization parameters α_ξ are independent of frequency, and for the case of the longitudinal Green's function the result is

$$\alpha_2 = \frac{1}{n_{02}}\left[\frac{2}{n_{02}}\lambda^L_2 + \frac{1}{n_{01}}\lambda^{T1}_2 + \frac{1}{n_{03}}\lambda^{T3}_2\right] - \beta\left[\left(\frac{n_0+n_2}{n^2_{02}} - 1\right)\lambda^L_1 + \left(\frac{n_0}{n_{01}n_{02}} - 1\right)\lambda^{T1}_1 + \left(\frac{n_0}{n_{03}n_{02}} - 1\right)\lambda^{T3}_1\right]. \tag{17}$$

The interaction is renormalized by

$$\gamma_2(i\omega_n) = \frac{1}{n^2_{02}}\left[2\lambda^L_1 - \left(n_0 + n_2 - n^2_{02}\right)K_2(i\omega_n)\right], \tag{18}$$

and the renormalized energy gap becomes frequency dependent:

$$\delta_2(i\omega_n) = \frac{1}{n_{02}}\left[\lambda^{T1}_1 + \lambda^{T3}_1 - \frac{1}{\beta}\sum_{n'} K_{31}(i\omega_{n'})\{n_{12}g_{12}(i\omega_{n'} - i\omega_n) + n_{23}g_{23}(i\omega_{n'} + i\omega_n)\}\right]. \tag{19}$$

The transverse Green's functions are determined in terms of the following renormalization parameters:

$$\alpha_1 = \frac{1}{n_{01}}\left[\frac{1}{n_{02}}\lambda_2^L + \frac{2}{n_{01}}\lambda_2^{T1} + \frac{1}{n_{03}}\lambda_2^{T3}\right] - \beta\left[\left(\frac{n_0}{n_{01}n_{02}} - 1\right)\lambda_1^L + \left(\frac{n_0+n_1}{n_{01}^2} - 1\right)\lambda_1^{T1} + \left(\frac{n_0}{n_{01}n_{03}} - 1\right)\lambda_1^{T3}\right],$$
$$\alpha_3 = \frac{1}{n_{03}}\left[\frac{1}{n_{02}}\lambda_2^L + \frac{1}{n_{01}}\lambda_2^{T1} + \frac{2}{n_{03}}\lambda_2^{T3}\right] - \beta\left[\left(\frac{n_0}{n_{03}n_{02}} - 1\right)\lambda_1^L + \left(\frac{n_0}{n_{01}n_{03}} - 1\right)\lambda_1^{T1} + \left(\frac{n_0+n_3}{n_{03}^2} - 1\right)\lambda_1^{T3}\right], \quad (20)$$

and, introducing a new frequency-dependent parameter $\zeta(i\omega_n)$,

$$\zeta(i\omega_n) = \lambda_1^{T1} + \lambda_1^{T3} - n_0 K_{31}(i\omega_n) - \frac{1}{\beta}\sum_{n'} K_{31}(i\omega_{n'})n_{13}g_{13}(i\omega_{n'} + i\omega_n), \quad \gamma_{31}(i\omega_n) = \frac{1}{n_{01}n_{03}}\zeta(i\omega_n) + K_{31}(i\omega_n). \quad (21)$$

Finally, the renormalization of the two energy gaps Δ_1 and Δ_3 are

$$\delta_1(i\omega_n) = \frac{1}{n_{01}}\left[\lambda_1^{T1} + \lambda_1^L - n_1 K_{31}(i\omega_n) + \frac{n_{13}}{n_{03}}\zeta(i\omega_n) - \frac{1}{\beta}\sum_{n'} K_2(i\omega_{n'})n_{12}g_{12}(i\omega_{n'} + i\omega_n)\right],$$
$$\delta_3(i\omega_n) = \frac{1}{n_{03}}\left[\lambda_1^{T3} + \lambda_1^L - n_3 K_{31}(i\omega_n) - \frac{n_{13}}{n_{01}}\zeta(i\omega_n) - \frac{1}{\beta}\sum_{n'} K_2(i\omega_{n'})n_{23}g_{23}(i\omega_{n'} + i\omega_n)\right]. \quad (22)$$

The renormalization parameters derived above account for all first-order terms appearing in the cumulant expansion of the local Green's functions. The small frequency-dependent sums, as for instance the last term in the expression for $\delta_2(i\omega_n)$, may to a good approximation be replaced by their values derived in the limit of $h \to 0$; i.e., $(1/\beta)\sum_{n'} K_p(i\omega_{n'})n_{12}g_{12}(i\omega_n - i\omega_{n'}) \approx K_p(i\omega_n)(n_1+n_2)/2$. The coupling parameters $K_p(i\omega_n)$ determining the effective medium are calculated by using Eq. (7), which calculation has to be carried out in a self-consistent way.

Defining the thermal expectation values $\langle a_{\xi\xi}\rangle \equiv \overline{n}_\xi$, the definition Eq. (3) implies that, for instance, $(1/\beta)\sum_n \mathcal{G}_2(i\omega_n) = -\overline{n}_0 - \overline{n}_2$. An analysis of the separate components of the Green's functions in Eq. (4) shows that

$$\overline{n}_0 - \overline{n}_\xi = \overline{n}_{0\xi} = \frac{n_{0\xi}}{1+\alpha_\xi} \quad (\xi = 1,2,3). \quad (23)$$

The exact relation $\overline{n}_0 + \overline{n}_1 + \overline{n}_2 + \overline{n}_3 = 1$ implies that the "sum rule" $(2/\beta)\sum_n \mathcal{G}_2(i\omega_n) = -1 - n_{02}/(1+\alpha_2)$ should be fulfilled at $h = 0$. As discussed in Ref. 16 the sum rules are, in principle, satisfied to the order considered in the cumulant expansion, but the generalization of the first-order results introduced by Eqs. (11)–(14) may introduce small violations of the sum rules. The simplest and most reliable results for $\overline{n}_\xi$ are obtained from Eq. (23) in combination with $\sum_\xi \overline{n}_\xi = 1$, which results have been used for calculating the moment per Cu^{2+} ion parallel to the applied field:

$$m_z = \frac{1}{N}\sum_i g\mu_B\langle S_z\rangle = \frac{1}{2}g\mu_b\overline{n}_{13}. \quad (24)$$

The present theory is valid in the paramagnetic phase, i.e., as long as the field is smaller than the critical one, $h < h_c$. Introducing the effective interaction parameters

$$\Lambda = J(\vec{Q}) - \gamma_{31}(0), \quad \overline{\Delta} = \Delta + \frac{\delta_1(0) + \delta_3(0)}{2}, \quad (25)$$

where $\vec{Q}$ is the ordering wave vector, the critical field is

$$h_c = \tfrac{1}{2}[\delta_1(0) - \delta_3(0) + \overline{n}_{13}\Lambda] + \left\{\overline{\Delta}[\overline{\Delta} - (\overline{n}_{01} + \overline{n}_{03})\Lambda] + \left[\tfrac{1}{2}\overline{n}_{13}\Lambda\right]^2\right\}^{1/2}. \quad (26)$$

III. THEORY IN COMPARISON WITH EXPERIMENTS

In the zero-temperature limit the present theory, to first order in $1/z$, predicts perfectly well-defined excitations in the paramagnetic phase, and, at zero field, the dispersion relation $E_{\vec{q}}$ for the dimer excitations is simplified into

$$E_{\vec{q}}^2 = (\Delta + 2\lambda_1)\left(\Delta + 2\lambda_1 - \frac{2}{1+4\lambda_2}[J(\vec{q}) - 2\lambda_1]\right), \quad (27)$$

where $\lambda_\nu = \lambda_\nu^L = \lambda_\nu^{T1} = \lambda_\nu^{T3}$ calculated from Eq. (15) or (16) at zero field and temperature. The combined results of Cavadini *et al.*[11] and Oosawa *et al.*[12] for the energies of the singlet-triplet

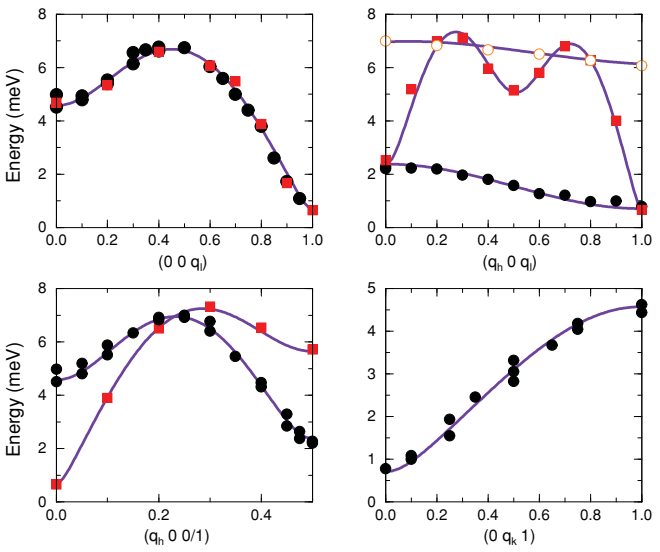

FIG. 1. (Color online) The dispersion relations for the singlet-triplet excitations in $TlCuCl_3$ at 1.5 K. The experimental results of Cavadini *et al.*[11] and Oosawa *et al.*[12] are shown by, respectively, black circles and red symbols (the precise values for the wave vectors in the different cases may be found by consulting the two papers). The solid lines are produced by the effective interaction parameters given by Eq. (28).

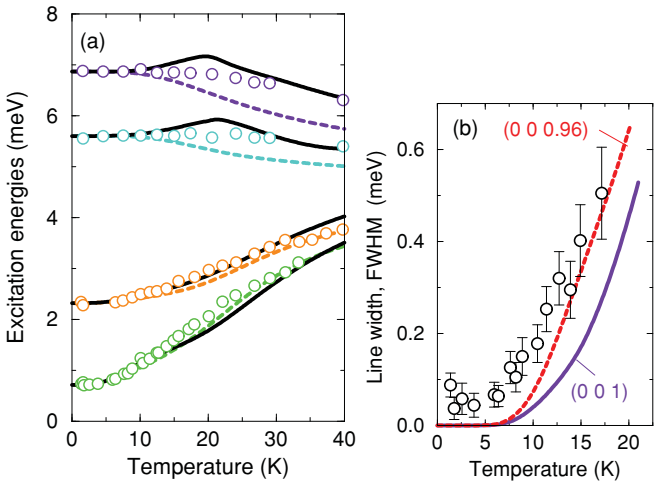

FIG. 2. (Color online) (a) The temperature dependencies of the energies of different dimer excitations at zero field. The experimental results are determined by Rüegg *et al.*[19] The solid lines are the predictions of the present theory. The dashed lines show for comparison the ScRPA results obtained, when using the model defined by Eq. (28). (b) The linewidth (FWHM) of the lowest excitation mode at zero field as a function of temperature. The circles are the experimental results obtained by Rüegg *et al.*[19] at (0 0 1) and (0 4 0). The solid line is the calculated result, and the dashed line shows the width predicted for the excitation at (0 0 0.96).

excitations in $TlCuCl_3$ at zero field and in the zero temperature limit are fitted by (in units of meV)

$$\begin{aligned} J_{\rm eff}(\vec{q}) = {} & 0.45\cos(\vec{q}\cdot\vec{a}) - 0.03\cos(2\vec{q}\cdot\vec{a}) \\ & + 1.56\cos[\vec{q}\cdot(2\vec{a}+\vec{c})] - 0.08\cos[\vec{q}\cdot(\vec{a}+\vec{c})] \\ & \mp\left\{1.03\cos\left[\vec{q}\cdot\left(\vec{a}+\tfrac{1}{2}\vec{c}\right)\right] - 0.13\cos\left(\tfrac{1}{2}\vec{q}\cdot\vec{c}\right)\right\} \\ & \times\cos\left(\tfrac{1}{2}\vec{q}\cdot\vec{b}\right) \end{aligned} \tag{28}$$

and $\Delta_{\rm eff} = 5.67$ meV, where $E_{\vec{q}}^2 = \Delta_{\rm eff}[\Delta_{\rm eff} - 2J_{\rm eff}(\vec{q})]$. The fit is shown in Fig. 1 and is the one used for determining Δ and $J(\vec{q})$ in Eq. (27).

The zero-field temperature dependencies of the energies and linewidths of a number of triplet excitations have been measured by Rüegg *et al.*[19] Their results for the excitation energies are compared with theory in Fig. 2(a). The experimental temperature dependence of the linewidth (FWHM) for the lowest-energy mode is shown in Fig. 2(b) in comparison with the calculated results for the excitation at (0 0 1) and the one lying nearby at (0 0 0.96). The rapid change of the linewidth shown by the modes close to the lower edge of the excitation band indicates that experimental resolution effects are important for this comparison. Furthermore, the calculated scattering intensities from the excitations are not symmetric around the intensity maxima, which leads to some arbitrariness in the assignments made for the calculated energies and linewidths. This uncertainty becomes important above 10 K, and, most pronouncedly, for the excitations belonging to the two upper modes in Fig. 2(a). This figure includes also the results obtained from the ScRPA theory. In order to enable a comparison of the two theories, the ScRPA results presented here are based on the model defined by Eq. (28) and not on the simplified one specified by Eq. (36) in Ref. 10. In addition to this, the high-temperature ScRPA results have been slightly improved by changing the power of the scale factor from 3 to 5 in the approximate expressions for $b_{xy}(\omega)$ and $b_z(\omega)$ in Eqs. (29) and (31) in Ref. 10.

The application of a field removes the degeneracy of the $S = 1$ triplet states of the dimers, and the collective singlet-triplet excitations split up into a longitudinal and a lower and an upper transverse mode, as shown in Fig. 3(a). The transition to the ordered phase takes place at the critical field H_c, where the energy of the lowest transverse mode vanishes. The calculated critical field as a function of temperature is compared with

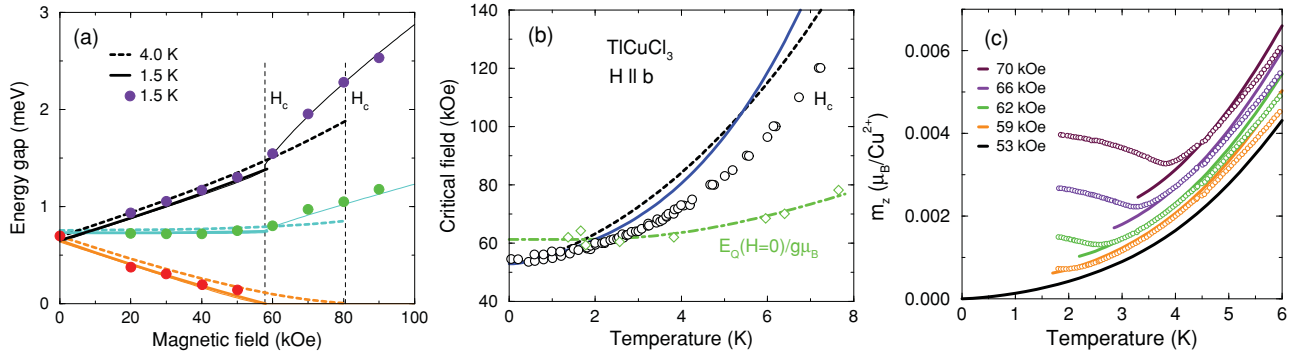

FIG. 3. (Color online) (a) The minimum energies of the three different dimer excitations as functions of a b-axis field. The solid circles are the experimental results obtained by Rüegg *et al.*[20,21] at 1.5 K. The thick lines show the predictions of the present $1/z$ theory at fields smaller than the critical field at the temperatures 1.5 K (solid) and 4.0 K (dashed). The thin lines are the results derived from the ScRPA theory in the ordered phase above H_c at 1.5 K. (b) The critical b-axis field in $TlCuCl_3$ as a function of temperature. The black circles show the experimental results obtained by Oosawa *et al.*[22] and Shindo and Tanaka.[23] The solid line is the behavior predicted to first order in $1/z$. Using the model defined by Eq. (28), the ScRPA theory produces the results shown by the dashed line. The diamonds are the experimental results of Rüegg *et al.*[19] for the zero-field energy of the soft mode divided by $g\mu_B$ (assuming $g = 2.06$). This would be the critical field (the dot-dashed line), if the renormalization effects were independent of field. (c) The parallel magnetic moment per Cu ion, m_z, as a function of temperature at various values of the field applied along the b direction. The system is predicted to stay disordered at all temperatures, when $H = 53$ kOe. The experimental points are a selection of those obtained by Oosawa *et al.*[1,6] The solid lines show the predictions of the present $1/z$ theory within the paramagnetic phase.

experiments in Fig. 3(b). The minute moment m_z induced by a constant field along the b axis has been calculated as a function of temperatures within the paramagnetic phase. The comparison of the calculated moment with experiments is shown in Fig. 3(c). The two Figs. 3(a) and 3(b) include the results obtained from the ScRPA theory presented above. In the case of Fig. 3(c), the ScRPA theory predicts a moment that at 6 K is about 10% larger than the $1/z$ results shown in the figure.

IV. CONCLUSION

The result in Sec. II for $\mathcal{G}_2(\vec{q},i\omega_n)$ simplifies into the Green's function for the singlet-singlet system derived in Ref. 16, if $K_{31}(i\omega_n)$ is assumed to be identical to zero. The present results are also consistent with those obtained in the singlet-doublet case.[14] The splitting of the triplet level caused by the field has here allowed a unique identification of all first-order terms in the expansion of the single-site Green's functions, and the present renormalization scheme is therefore more trustworthy than the one applied in Ref. 14.

The two theories, the ScRPA theory developed in Ref. 10 and the present diagrammatic expansion of the Green's functions to first order in $1/z$, are closely related, and their predictions for the paramagnetic properties of $TlCuCl_3$ show only minor differences between each other. In both cases, the final results are, in principle, determined alone by the model parameters. In reality, some approximations are required in order to carry out the numerical analyses. Those made in the present theory have no consequences for the results (as long as $h \ll \Delta$), whereas the approximations made for $b_{xy}(\omega)$ and $b_z(\omega)$ in Ref. 10 are less transparent. One consequence of the approximations made in Ref. 10 is that all linewidth effects are neglected. This may be a fair approximation below 10 K, but the linewidths are important for characterizing the excitation spectra above 10 K, and it is encouraging that the $1/z$ theory accounts reasonably well for the temperature dependence of the linewidths observed experimentally[20] at zero field.

The model used in the present calculations is in complete accord with the experimental dispersion relations obtained at zero field and temperature (1.5 K), whereas the simplified model used in Ref. 10 leads to a smaller density of states just above the lower threshold energy. The calculated values of the critical field and the magnetization close to the critical field, Figs. 3(b) and 3(c), react extremely sensitively to this minor change of the model, as indicated by the comparison of the ScRPA results shown here and those presented in Ref. 10. This strong sensitivity may just reflect the breakdown of any perturbation theory close to the singular point of a phase transition. However, it is apparent that both theories overestimate the self-consistent enhancement of the renormalization effects, when the critical field is approached, and that this critical enhancement needs to be moderated by contributions of higher order. It is worth noting that this shortcoming of the theory increases with temperature—that it is the thermal rather than the quantum critical fluctuations which are the cause of the discrepancies.

The ScRPA theory may be considered to be less rigorous than the present theory. On the other hand, the simplicity of the ScRPA theory made it possible to extend it to the case of the ordered phase appearing above the critical field.[10] The good agreement obtained here, between the predictions of the two theories in the paramagnetic phase, indicates that the ScRPA results obtained for the ordered phase may also be trusted. This is a valuable conclusion, since the more stringent $1/z$ expansion of the Green's functions is going to be extremely complicated to apply in the case of the ordered phase, where the number of interaction terms in the Hamiltonian is doubled.

ACKNOWLEDGMENTS

The author would like to thank Henrik Smith and Christian Rüegg for stimulating discussions and useful comments.

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