Magnon Dispersion in the Field-Induced Magnetically Ordered Phase of $TlCuCl_3$

Masashige Matsumoto,[1,*] B. Normand,[2] T. M. Rice,[1] and Manfred Sigrist[1]
[1]*Theoretische Physik, ETH-Hönggerberg, CH-8093 Zürich, Switzerland*
[2]*Département de Physique, Université de Fribourg, CH-1700 Fribourg, Switzerland*
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The magnetic properties of the interacting dimer system $TlCuCl_3$ are investigated within a bond-operator formulation. The observed field-induced staggered magnetic order perpendicular to the field is described as a Bose condensation of magnons which are linear combinations of dimer singlet and triplet modes. This technique accounts for the magnetization curve and for the field dependence of the magnon dispersion curves observed by high-field neutron scattering measurements.

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$TlCuCl_3$ is an insulating quantum spin system with a gap in the spin excitation spectrum [1] at zero field which originates from dimerization of the $S = 1/2$ spins of the Cu^{2+} ions. The compound is isostructural to $KCuCl_3$ and the crystal structure can be considered as coupled two-leg ladders separated by Tl^+ ions. However, inelastic neutron scattering (INS) measurements of triplet magnon excitations found that the magnon modes have significant dispersion in all three spatial dimensions for both $TlCuCl_3$ and $KCuCl_3$ [2–4]. This indicates that these compounds are three-dimensional (3D) interacting dimer systems with interladder interactions stronger than the interdimer interactions within the ladders. As observed in INS experiments by Cavadini *et al.*, the magnon modes are split into three by a magnetic field, with splitting proportional to the field, and the lowest mode becomes soft at a critical field H_c [5]. For $TlCuCl_3$ ($KCuCl_3$), the zero-field excitation gap and H_c are, respectively, 0.7 meV (2.6 meV) and 5.7 T (20 T).

For $H > H_c$, both compounds show a uniform magnetization parallel to the field [6]. Elastic neutron scattering measurements on $TlCuCl_3$ show in addition a staggered magnetic order perpendicular to the field [7]. A Goldstone mode is then expected due to the breaking of rotational symmetry around the field axis. Very recently, Rüegg *et al.* observed such a gapless mode for $H > H_c$ in $TlCuCl_3$ [8], and also reported a renormalized field dependence of the higher magnon modes [9].

Field-induced magnetic order in otherwise gapped ladder systems has been described [10,11] by theoretical approaches which focus only on the singlet and the lowest triplet magnon. Giamarchi and Tsvelik [12] cast their theory as a Bose condensation of a soft mode, and for $TlCuCl_3$ the Bose condensation of magnons has been used [13,14] to account for the observed temperature dependence of the magnetization. A quantum Monte Carlo simulation on a simplified 3D cubic lattice was also in agreement with an interpretation as magnon condensation [15].

We note that the structure of the observed staggered order is related to the wave vector of the soft magnon [5,7], in support of the idea of a magnon Bose condensation. In this paper we develop a microscopic theory of field-induced magnetic order which takes into account the higher triplet modes. These modes, as we will show below, must be included to obtain a complete description of the condensate and of the evolution of the magnon dispersion in the presence of field-induced magnetic order. We use a bond-operator formulation which retains all four states of each dimer, analogous to the treatment of bilayer antiferromagnets by Sommer *et al.* [16].

For the parametrization of the couplings between the Cu^{2+} ion spins, we follow Ref. [3]. The unit cell contains two equivalent dimers, and the Hamiltonian is given by

$$\mathcal{H} = \sum_j \Big[J \boldsymbol{S}^1_{l,j} \cdot \boldsymbol{S}^1_{r,j} + J'_2 \boldsymbol{S}^1_{l,j} \cdot \boldsymbol{S}^1_{r,j+d_2} + J_1 \sum_{n=l,r} \boldsymbol{S}^1_{n,j} \cdot \boldsymbol{S}^1_{n,j+d_1} + J'_1 \boldsymbol{S}^1_{l,j} \cdot \boldsymbol{S}^1_{r,j+d_1} + J_3 (\boldsymbol{S}^1_{l,j} \cdot \boldsymbol{S}^2_{l,j+d_{3+}} + \boldsymbol{S}^1_{r,j} \cdot \boldsymbol{S}^2_{r,j+d_{3-}}) + J'_3 (\boldsymbol{S}^1_{l,j} \cdot \boldsymbol{S}^2_{r,j+d_{3+}} + \boldsymbol{S}^1_{r,j} \cdot \boldsymbol{S}^2_{l,j+d_{3-}}) \Big] + [1 \leftrightarrow 2]. \quad (1)$$

Here $\boldsymbol{S}^m_{n,j}$ is the spin $S = 1/2$ operator in unit cell j on the sublattice $m = 1, 2$, and n ($= l, r$) denotes the left or right spin of the dimer. $\boldsymbol{d}_1 = \hat{\boldsymbol{a}}$, $\boldsymbol{d}_2 = 2\hat{\boldsymbol{a}} + \hat{\boldsymbol{c}}$, and $\boldsymbol{d}_{3\pm} = \hat{\boldsymbol{b}}/2 \pm (\hat{\boldsymbol{a}} + \hat{\boldsymbol{c}}/2)$, where $\hat{\boldsymbol{a}}$, $\hat{\boldsymbol{b}}$, and $\hat{\boldsymbol{c}}$ are unit vectors corresponding to the a, b, and c axes, respectively [3].

Because the intradimer exchange coupling J is the largest [3,4], we introduce bond operators $s^\dagger$ and $t^\dagger_\alpha$ based on these dimers [17]. In the presence of an external field, the appropriate operators are [18]

$$s^\dagger|0\rangle = (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)/\sqrt{2}, \qquad t^\dagger_+|0\rangle = -|\uparrow\uparrow\rangle,$$
$$t^\dagger_0|0\rangle = (|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)/\sqrt{2}, \qquad t^\dagger_-|0\rangle = |\downarrow\downarrow\rangle.$$

These have Bose statistics and are subject to the constraint

$s^\dagger s + \sum_\alpha t_\alpha^\dagger t_\alpha = 1$ at each dimer $\{j, m\}$, where $\alpha = +, 0, -$. We introduce the Fourier transformation $c_j^{m\dagger} = (1/N)\sum_{\mathrm{k}} c_{\mathrm{k}}^{m\dagger} e^{i\mathbf{k}\cdot\mathbf{r}_j^m}$ for the dimer operators, where $2N$ is the total dimer number.

We restrict our considerations to zero temperature, and begin with the low-field region ($H \le H_c$). Here the dimer singlets have the lowest energy, so the s bosons are taken to be condensed. The s operators are replaced by a c number $s_{\boldsymbol{k}}^m = \bar{s}_0 \delta_{\boldsymbol{k},0}$ [19], and the local constraint relaxed to the global one $\bar{s}_0^* \bar{s}_0 = N - \sum_{\boldsymbol{k},\alpha} t_{\boldsymbol{k}\alpha}^{m\dagger} t_{\boldsymbol{k}\alpha}^m$. Retaining quadratic terms in the triplet operators,

$$\mathcal{H} = \sum_{m,\mathrm{k}} \Big\{ \sum_\alpha [(J - \alpha g \mu_{\mathrm{B}} H + f_{\mathbf{k}}) t_{\mathrm{k}\alpha}^{m\dagger} t_{\mathrm{k}\alpha}^m + g_{\boldsymbol{k}} t_{\boldsymbol{k}\alpha}^{m\dagger} t_{\boldsymbol{k}\alpha}^{\bar{m}}] + \sum_{\alpha=+,-} (f_{\boldsymbol{k}} t_{\boldsymbol{k}\alpha}^m t_{-\boldsymbol{k}\bar{\alpha}}^m + g_{\boldsymbol{k}} t_{\boldsymbol{k}\alpha}^m t_{-\boldsymbol{k}\bar{\alpha}}^{\bar{m}} + \mathrm{H.c.}) + (f_{\boldsymbol{k}} t_{\boldsymbol{k}0}^m t_{-\boldsymbol{k}0}^m + g_{\boldsymbol{k}} t_{\boldsymbol{k}0}^m t_{-\boldsymbol{k}0}^{\bar{m}} + \mathrm{H.c.})/2 \Big\}, \quad (2)$$

where $f_{\boldsymbol{k}} = J_a \cos k_x + J_{a2c} \cos(2k_x + k_z)$, $g_{\boldsymbol{k}} = 2J_{abc} \cos(k_x + k_z/2)\cos(k_y/2)$, $\bar{\alpha} = -\alpha$, and $\bar{m} = 2, 1$ for $m = 1, 2$. The effective interdimer interactions [3] are given by $J_a = J_1 - J_1'/2$, $J_{a2c} = -J_2'/2$, and $J_{abc} = (J_3 - J_3')/2$, where we note changes in the signs of the parameters J_{a2c} and, for $J_1' > 2J_1, J_a$. Introducing the operators $t_{\boldsymbol{k}\alpha}^{\pm} = (t_{\boldsymbol{k}\alpha}^1 \pm t_{\boldsymbol{k}\alpha}^2)/\sqrt{2}$ gives two independent ($\pm$) modes [20] whose eigenvalues are obtained by the Bogoliubov transformation [18]

$$E_{\boldsymbol{k}\alpha}^{\pm} = \sqrt{(J + f_{\boldsymbol{k}} \pm g_{\boldsymbol{k}})^2 - (f_{\boldsymbol{k}} \pm g_{\boldsymbol{k}})^2} - \alpha g \mu_{\mathrm{B}} H. \quad (3)$$

The Brillouin zone lies between $-\pi$ and π (unit lattice spacing) in each direction, and the two branches correspond to the two-sublattice system. We treat only the + mode in the expanded Brillouin zone ($-2\pi \le k_z \le 2\pi$) in the z direction, because the magnon dispersions obey the relations $E_{\boldsymbol{k}\alpha}^- = E_{\mathrm{k}\pm(0,0,2\pi),\alpha}^+$. We extract the effective interactions (J, J_a, J_{a2c}, J_{abc}) from the data at zero field for both $TlCuCl_3$ and $KCuCl_3$ (Fig. 1), and these are listed in Table I. The values are consistent with the results of Ref. [4]. They are also similar to those of Ref. [3], but not identical because the current treatment [leading to (3)] goes beyond a pure dimer description. The signs are consistent with the expectation that all intersite parameters $\{J\}$ in (1) are antiferromagnetic.

The three magnon modes are degenerate in zero field, and, as shown in Fig. 2, are split linearly by an external field in agreement with INS results [5]. Below H_c, a $t_+^\dagger$ triplet excited from the singlet condensate may propagate due to the interaction between triplets and singlets. The wave function of this excited state can be approximated by a linear combination of dimer singlets and triplets as

$$|\psi\rangle \sim u(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle) - v e^{i(\boldsymbol{k}\cdot\boldsymbol{r}_j^m - E_{k+}t)} |\uparrow\uparrow\rangle, \quad (4)$$

where u is of order unity and v is a small, real coefficient. The expectation values of the spin operator components at a given dimer j, m in the state (4) are

$$\begin{aligned} \langle S_{l,z}\rangle_{j,m} &= \langle S_{r,z}\rangle_{j,m} \sim v^2/2, \\ \langle S_{l,x}\rangle_{j,m} &= -\langle S_{r,x}\rangle_{j,m} \sim (uv/2)\cos(\boldsymbol{k}\cdot\boldsymbol{r}_j^m - E_{\boldsymbol{k}+}t), \\ \langle S_{l,y}\rangle_{j,m} &= -\langle S_{r,y}\rangle_{j,m} \sim -(uv/2)\sin(\boldsymbol{k}\cdot\boldsymbol{r}_j^m - E_{\boldsymbol{k}+}t). \end{aligned} \quad (5)$$

This excited mode has a very small, uniform magnetic moment parallel to the field, and thus gains Zeeman energy. Perpendicular to the field, it also possesses a finite magnetic moment, which is characterized by the wave vector $\boldsymbol{k}$ and energy $E_{\boldsymbol{k},+}^+$, and is staggered (spins l and r oppositely aligned) due to the antiferromagnetic intradimer coupling. For the mode $t_-^\dagger$, the direction of the uniform magnetic moment is antiparallel to the field, leading to a higher Zeeman energy. Finally, the $t_0^\dagger$ mode has no magnetization perpendicular to the field, and a moment parallel to the field which is modulated with wave vector $\boldsymbol{k}$, so its energy does not shift with the field. On increasing the field, these modes shift position without changing the shape of their dispersion (3), and the lowest ($\alpha = +$)

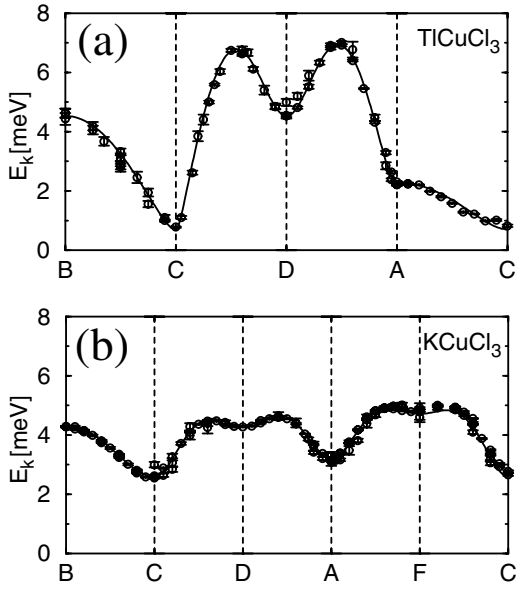

FIG. 1. Zero-field magnon dispersion relations for $TlCuCl_3$ and $KCuCl_3$. The x-axis labels represent the reciprocal-space points $B = (0, 2\pi, 2\pi)$, $C = (0, 0, 2\pi)$, $D = (0, 0, 0)$, $A = (\pi, 0, 0)$, and $F = (\pi, 0, 2\pi)$. Experimental results [3] are shown as points, and were measured at $T = 1.5$ K (5 K) for $TlCuCl_3$ ($KCuCl_3$). Solid lines are theoretical results using the parameters listed in Table I.

TABLE I. Effective dimer interactions.

	$TlCuCl_3$	$KCuCl_3$
J [meV]	5.501	4.221
J_a [meV]	-0.215	-0.212
J_{a2c} [meV]	-1.581	-0.395
J_{abc} [meV]	0.455	0.352

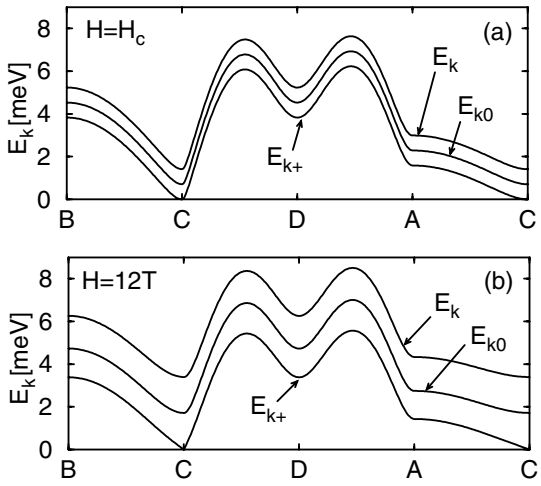

FIG. 2. Calculated magnon dispersions for $TlCuCl_3$ at $H = H_c(= 5.6\ \mathrm{T})$ (a), and $H > H_c$ ($H = 12$ T) (b).

mode becomes gapless at the point C with $\boldsymbol{Q} \equiv (0, 0, 2\pi)$ [Fig. 2(a)], which determines the critical field. Thus at $H = H_c$, the lowest mode exhibits a quadratic dependence on $\boldsymbol{k}$ around $\boldsymbol{Q}$.

To describe the field regime with $H > H_c$, the Hamiltonian (2) must be extended to include triplet-triplet interactions. The coefficients of these terms involve combinations of the intersite exchange constants in (1) beyond the interdimer interactions in (2). To determine these we have made the simplifying assumption that $J_1 = J_3' = 0$, so that the three remaining coefficients, J_1', J_2', and J_3, are specified by the interdimer interactions. Because the additional triplet-triplet interactions are largely governed by terms involving J_2', this is not a significant approximation. A further assumption is made by neglecting terms involving three t operators, which give only small corrections concentrated in the region of maximum staggered magnetic order. For $H > H_c$, Bose condensation of the lowest triplet implies a macroscopic occupation of the $t_{\boldsymbol{k}+}$ mode at $\boldsymbol{Q}$.

For a full description of this regime, the singlet and triplet operators are transformed [16] to

$$\begin{aligned} a^m_{\boldsymbol{k}} &= u s^m_{\boldsymbol{k}} + v(x t^m_{\boldsymbol{k}+\boldsymbol{Q},+} + y t^m_{\boldsymbol{k}+\boldsymbol{Q},-}), \\ b^m_{\boldsymbol{k}+} &= u(x t^m_{\boldsymbol{k}+} + y t^m_{\boldsymbol{k}-}) - v s^m_{\boldsymbol{k}+\boldsymbol{Q}}, \qquad b^m_{\boldsymbol{k}0} = t^m_{\boldsymbol{k}0}, \\ b^m_{\boldsymbol{k}-} &= x t^m_{\boldsymbol{k}-} - y t^m_{\boldsymbol{k}+}. \end{aligned} \tag{6}$$

The $\boldsymbol{k}$-independent coefficients u, v, x, and y arise from two unitary transformations, and may be written as $u = \cos\theta$, $v = \sin\theta$, $x = \cos\phi$, $y = \sin\phi$, with θ and ϕ to be determined. We treat the $a^m_{\boldsymbol{k}}$ operator as uniformly condensed, $a^m_{\boldsymbol{k}} = \bar{a}_0 \delta_{\boldsymbol{k},0}$, and the ground state as a coherent condensate of the a^m_0 operator. We emphasize that the highest triplet mode ($t^m_{\boldsymbol{Q}-}$) also participates in the condensate, because $\mathcal{H}$ contains processes $t^\dagger_+ t^\dagger_- ss$ which nucleate t_+ and t_- triplets from singlets. The linear combination of singlet and triplets in the condensate a^m_0 yields a staggered magnetization perpendicular to the field with wave vector $\boldsymbol{Q}$, as observed in Ref. [7].

With the transformation of Eq. (7), the Hamiltonian to quadratic order in the b operators takes the form $\mathcal{H} = O(b^0) + O(b^1) + O(b^2)$. Because the particle number is unaltered, the c-number $\bar{a}_0$ may be replaced by $\bar{a}^*_0 \bar{a}_0 = N - \sum_{\boldsymbol{k},\alpha} b^{m\dagger}_{\boldsymbol{k}\alpha} b^m_{\boldsymbol{k}\alpha}$. The constant terms ($O(b^0)$) represent the mean-field energy of the a_0 condensate, and the parameters (θ, ϕ) are chosen to minimize this energy, which also eliminates the $O(b^1)$ terms in the transformed Hamiltonian. The critical field H_c is determined by the condition $\theta \to 0$, which gives purely singlet character to the condensate. The limit ($\theta = \pi/2$, $\phi = 0$) gives a condensate with purely triplet t_+ character, and determines the saturation field H_s for full parallel polarization. The values for H_c and H_s are

$$\begin{aligned} H_c &= \sqrt{J^2 - 2J(|J_a| + |J_{a2c}| + 2J_{abc})}/(g\mu_{\mathrm{B}}), \\ H_s &= (J + 2|J_a| + 2|J_{a2c}| + 4J_{abc})/(g\mu_{\mathrm{B}}), \end{aligned} \tag{7}$$

where the form of H_c coincides with the soft-mode condition in the low-field regime.

The $O(b^2)$ terms are diagonalized by a Bogoliubov transformation which yields the energies of the collective modes of the condensate. Above H_c, the lowest mode remains gapless, but develops a linear dependence on $\boldsymbol{k}$ near $\boldsymbol{Q}$ [Fig. 2(b)]. This is a Goldstone mode: a staggered magnetic moment $M_\perp$, whose mean-field value is $M_\perp = uv(x + y)/\sqrt{2}$, is induced perpendicular to the magnetic field, and this breaks rotational symmetry around the axis parallel to the field. Rotations of this induced staggered moment are realized by changing the phase of x and y in Eq. (7) ($x \to e^{-i\chi}x$, $y \to e^{i\chi}y$, where χ is the rotation angle), and do not change the energy. As a result, the Goldstone mode remains gapless, and the field dependence of the higher modes is also renormalized, in agreement with experiment (Fig. 3). The energy gaps of the higher modes E_{g0} and E_{g-} show an abrupt increase in slope at H_c (Fig. 3). At the saturation field H_s, the $\boldsymbol{k}$ dependence of the lowest excitation mode near $\boldsymbol{Q}$ becomes quadratic again. In the high-field region above H_s, the condensate consists only of the lowest-lying triplet, and a gap reopens in the spectrum of the lowest (pure singlet) excitation mode (inset of Fig. 3).

We may also consider the parallel and perpendicular magnetization curves. For $TlCuCl_3$ ($KCuCl_3$), the parameters of Table I give a critical field of $H_c = 5.6$ T (19.6 T), which is consistent with the measured value [6]. For both $TlCuCl_3$ and $KCuCl_3$, the square of the staggered moment ($M^2_\perp$) has linear dependence close to H_c and to H_s (Fig. 4), indicating that $M_\perp \sim \sqrt{H - H_c}$. For $KCuCl_3$, the magnetization parallel to the field ($M_\parallel$) is almost linear in H, as shown in Fig. 4(a), in good agreement with experiment. For $TlCuCl_3$, the field dependence of $M_\parallel$ appears not to be linear in H, again in very good agreement with the observed form [6]. The theoretical mean-field value is

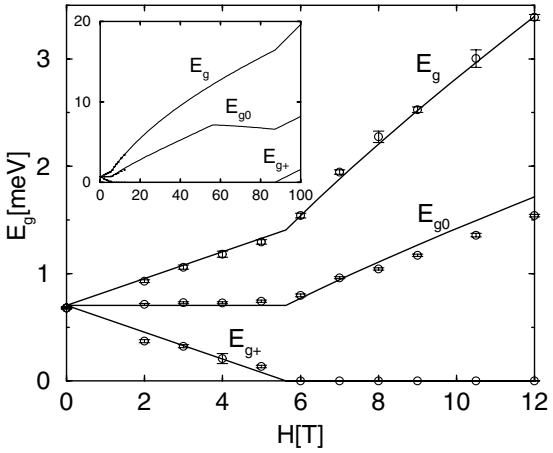

FIG. 3. Field dependence of the energy gap of the three magnon modes in $TlCuCl_3$. Points are data from the INS experiment of Ref. [9], and solid lines the results of the present theory. Inset: field dependence at high fields. We have used $g = 2.16$ for the g factor of $TlCuCl_3$ [9].

given by $M_{\|} = v^2(x^2 - y^2)$, from which it is clear that this difference is due to the magnitude of the interdimer interactions. If the contribution to the a_0^m operator of the highest triplet mode (t_-) is neglected [Eq. (7)], $M_{\|}$ would become completely linear in H. However, the strong interdimer interactions involve the highest mode largely through interactions of the type $t_+^\dagger t_-^\dagger ss$, which cost Zeeman energy, thus reducing the value of v for the a_0^m operator, and consequently $M_{\|}$ is suppressed near H_c for $TlCuCl_3$.

We have studied the evolution of the magnon dispersion in $TlCuCl_3$ and $KCuCl_3$ as the magnetic field is tuned through the quantum critical point separating a gapped spin-liquid state from a state of field-induced staggered magnetic order which exists between the critical and saturation fields. A consistent theory requires the admixture of both the lowest and highest triplet modes into the singlet dimer state to form a Bose condensate. The spectrum has a gapless Goldstone mode associated with the breaking of rotational symmetry by the staggered magnetic order, as observed by Rüegg *et al.* [8] for $TlCuCl_3$. The two higher excitation modes are also renormalized, in further agreement with observation [9]. Finally, our zero-temperature mean-field description is also in good accord with measurements [6,7,21] of the uniform and staggered magnetization for both $KCuCl_3$ and $TlCuCl_3$.

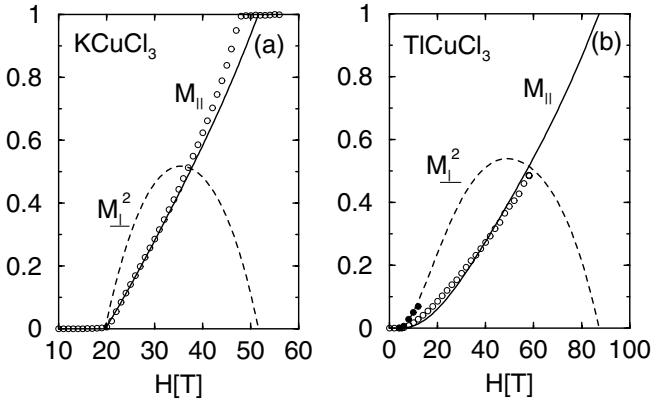

FIG. 4. Normalized magnetization curves for $KCuCl_3$ (a), and $TlCuCl_3$ (b). The solid line ($M_{\|}$) is the magnetization parallel to the field and the dashed line ($M_{\|}^2$) the square of the magnetization perpendicular to the field. Open (filled) points are experimental data for $M_{\|}$ ($M_{\perp}^2$) measured at $T = 1.3$ K (0.2 K) [7,21], where $g = 2.29$ is the g factor for $KCuCl_3$. For $TlCuCl_3$ we have used the same g as in Fig. 3.

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*Department of Physics, Faculty of Science, Shizuoka University, 836 Oya, Shizuoka 422-8529, Japan.

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