Appl. Phys. A 74 [Suppl.], S840–S842 (2002) / Digital Object Identifier (DOI) 10.1007/s003390101156

Applied Physics A
Materials
Science & Processing

Spin dynamics in the high-field phase of quantum-critical $S = 1/2$ $TlCuCl_3$

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Received: 18 July 2001/Accepted: 24 October 2001 –

Abstract. An external magnetic field suppresses the spin-energy gap in singlet ground state $S = 1/2$ $TlCuCl_3$. The system becomes quantum-critical at $H_c \approx 5.7\,T$, where the energy of the lowest Zeeman-split triplet excitation crosses the nonmagnetic ground state. Antiferromagnetic ordering is reported above H_c, which underlines the three-dimensional nature of the observed quantum phase transition. The intrinsic parameters of $S = 1/2$ $TlCuCl_3$ allow us to access the critical region microscopically by neutron scattering. A substantial study of the spin dynamics in the high-field phase of $TlCuCl_3$ at $T = 1.5\,K$ up to $H = 12\,T$ was performed for the first time. The results possibly indicate two dynamical regimes, which can be understood within characteristically renormalized triplet modes and a low-lying dynamics of potentially collective origin.

PACS: 75.10.Jm; 75.40.Gb; 78.70.Nx

The phase diagram of quantum magnets and the nature of the elementary states around the quantum-critical points, defined by pressure, doping or external magnetic fields, recently attracted a considerable theoretical and experimental interest. For these zero-temperature phase transitions in the quasi-one-dimensional case a substantial experimental survey is presented by Chaboussant et al. [1], where in particular the $S = 1/2$ Heisenberg system $Cu_2(C_5H_{12}N_2)_2Cl_4$ is explained in the gapless phase by a mapping of strongly coupled spin ladders in a magnetic field on the exactly solvable XXZ model. But dynamical studies of higher-dimensional systems in (Q, ω) by inelastic neutron scattering are lacking to date. $TlCuCl_3$ crystallizes in the space group $P2_1/c$ and is well characterized as a three-dimensional quantum magnet in the dimer limit, with a nonmagnetic ground state and a finite energy gap to elementary triplet waves, which show dispersive behavior in all directions of the reciprocal space [2]. The modest spin gap $\Delta \approx 0.7\,meV$ in $TlCuCl_3$ and the availability of large single crystals give the unique possibility to investigate a quantum phase transition in a broad range of the physically relevant parameters like temperature and magnetic field. In $TlCuCl_3$, the Zeeman splitting of the triplet modes, similarly to isostructural $KCuCl_3$ [3, 4], reduces the gap at the minimum of the energy dispersion and the lowest-lying branch $|1,+1\rangle$ can energetically fall below the value of the nonmagnetic singlet ground state, see Fig. 1.

Subject to the condition that the quantum phase transition is of three-dimensional nature, at the critical field H_c magnetic long-range ordering is expected because each dimer unit is preferably in a renormalized triplet state $|1,+1\rangle'$ with a finite net moment and the three-dimensional couplings are provided by the inter-dimer magnetic exchange. The magnetization mixes the initially pure eigenstates of the system to the renormalized states, which have to be determined in order to describe the resulting excitations in the quantum-critical regime. The accessible field strength for inelastic neutron-scattering investigations is currently limited to $H_{max} = 14.5\,T$, which drastically reduces the number of quantum-critical compounds with $\Delta/g\mu_B < H_{max}$. But for $TlCuCl_3$ the condition is fulfilled. The above-mentioned

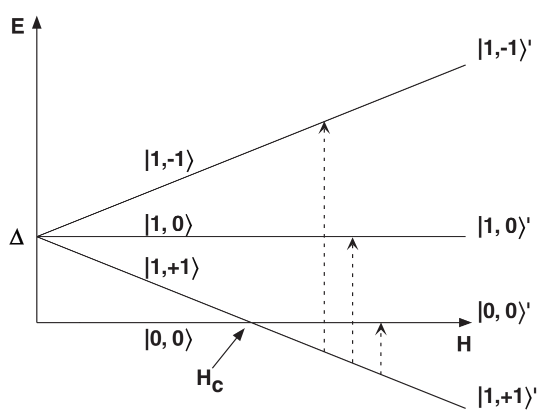

Fig. 1. Schematic representation of the expected quantum phase transition in $TlCuCl_3$ at $H_c \approx 5.7$ T. Above H_c the initial eigenstates of the system are mixed by the field-induced long-range ordering to the primed states. For clarity, the corresponding energy renormalization in the high-field phase is not shown

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magnetic ordering for $H > H_c$ was recently investigated by Tanaka et al. [5]. A theory by Nikuni et al. [6] based on Bose–Einstein condensation (BEC) of dilute magnons can explain the data of the neutron-diffraction study and the magnetization measurements presented in the cited literature. The spin arrangement was found to be perpendicular to the applied field direction, with a staggered magnetization $m = 0.26\,\mu_B$ per Cu^{2+} ion at $H = 12$ T. This orientation is predicted theoretically by Tachiki and Yamada [7]. The quantum-critical phase boundary in $TlCuCl_3$ is well described by a power law derived form the BEC theory. However the critical exponent given by the theory $\phi_{BEC} = 1.5$ is slightly smaller than the observed value $\phi_{obs} = 2.0(1)$ [5], which motivates additional microscopic investigations.

1 Measurements

A cylindrical single crystal of $TlCuCl_3$ was mounted in the VM1 cryomagnet on the V2 three-axis spectrometer for cold neutrons FLEX (HMI, Berlin). The orientation of the sample was determined by the scattering plane spanned by the $(0,2,0)$ and $(1,0,4)$ nuclear Bragg reflections. Measurements were performed at $T = 1.5$ K and vertical fields $0 \leq H \leq 12$ T, in constant final energy mode $E_f = 4.7$ meV with a cold Be filter in front of the analyzer. The parameters of the horizontally focussing monochromator, the flat analyzer and the chosen 60–60–60 collimation conditions were implemented in the Monte Carlo program RESTRAX4 to simulate the expected peak form depending on the energy transfers up to 4 meV. The highly asymmetric line shapes were taken into account in the analysis of the inelastic scans. Considering the intensity modulation by the dimer structure factor in the scattering plane, spectra of the Zeeman-split triplet excitations were collected at the minimum point $Q = (0,4,0)$ reciprocal lattice units (r.l.u.) of the dispersion below the quantum-critical value $H_c \approx 5.7$ T to complete earlier investigations performed on TASP (SINQ PSI, Villigen) between zero field and $H = 4$ T. The spin dynamics in the quantum-critical phase of $TlCuCl_3$ was successively investigated at the same point above H_c. The observed inelastic neutron profiles show two sharp transitions up to $H = 12$ T, where additionally a low-lying signal emerges from the elastic flank. In Fig. 2 three examples of the high-field phase measurements are presented. The refinement based on a full 4D convolution of sharp excitations well reproduces the observations. The $|1,+1\rangle' \rightarrow |1,-1\rangle'$ neutron intensity remains stable compared to $|0,0\rangle \rightarrow |1,-1\rangle$ in the Zeeman regime, whereas the $|1,+1\rangle' \rightarrow |1,0\rangle'$ transition is slightly more pronounced in the high-field phase than the corresponding $|0,0\rangle' \rightarrow |1,0\rangle$ excitation for $H < H_c$.

The disposable data set of the field dependence in $TlCuCl_3$ is summarized in Fig. 3, including the energy information of all observed transitions as well as the earlier TASP data to complete the Zeeman-split excitations in the undercritical regime.

The dashed lines in Fig. 3 are a fit to the data using a simple linear model with the following parameters. In the Zeeman regime, the spin-gap energy $\Delta = 0.71(1)$ meV and the two field-dependent $\Delta S^z = \pm 1$ modes split by the fitted gyromagnetic factor $g = 2.16(5)$ agree well with the observation, consistently with the values given by Oosawa et al. [8].

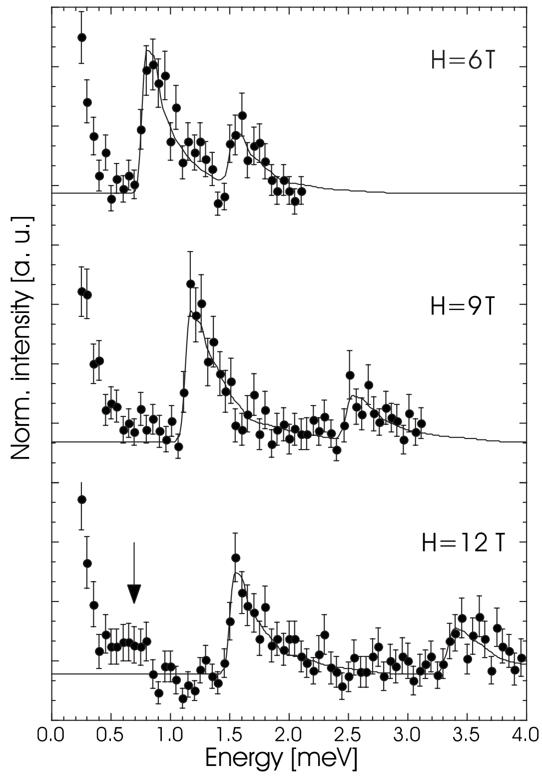

Fig. 2. Observed spectra in $TlCuCl_3$ at $T = 1.5$ K. At the highest magnetic field a weak signal appears at the flank of the elastic line (*arrow*), whose origin is addressed in the text

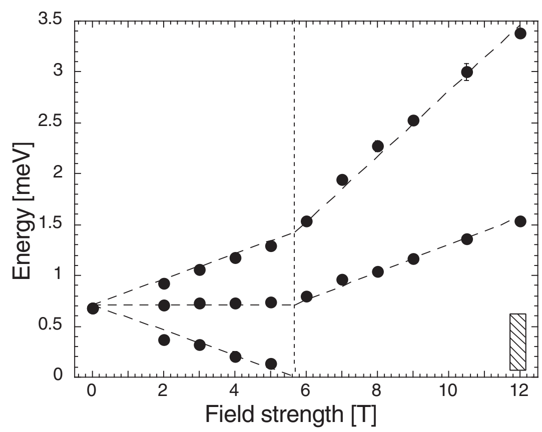

Fig. 3. Energies of the observed transitions for the Zeeman regime and for the high-field phase in $TlCuCl_3$ at $T = 1.5$ K. The characteristics are explained in the text

The $\Delta S^z = 0$ mode remains unshifted. Extrapolation yields $H_c = 5.7(1)$ T, which is in excellent agreement with earlier macroscopic measurements [8]. In the high-field phase above H_c the two sharp transitions show a Zeeman-like behavior with energies following linear relations

$$E_{|1,0\rangle'}(H) = E_{|1,0\rangle}(H_c) + g'\mu_B(H - H_c), \quad (1)$$

$$E_{|1,-1\rangle'}(H) = E_{|1,-1\rangle}(H_c) + g''\mu_B(H - H_c), \quad (2)$$

with $g'' = 5.58(21)$ and $g' = 2.36(8)$. Fitting the isolated dimer model according to the simple picture presented in Fig. 1, $g'' = 2g'$ with $g' = 2.50(7)$ can not account for the observed features. The low-lying signal at $H = 12\,\mathrm{T}$ is indicated by a hatched rectangle and represents the only evidence found for a possible $|1, +1\rangle' \to |0, 0\rangle'$ transition.

2 Discussion

The obtained values for g' and g'' clearly indicate that a simple isolated dimer model with linear field dependences of the transition energies becomes less accurate above the quantum-critical point, as expected considering the reported three-dimensional order. Regarding the main transitions, a model based on perturbed dimer states is introduced, which qualitatively explains the two sharp modes observed in the high-field phase of $TlCuCl_3$. The effective Hamiltonian reads

$$H = H_\mathrm{D} + H_\mathrm{Z} + H_\mathrm{AF} + H_\mathrm{F}, \tag{3}$$

where H_D denotes the dimer interaction term, H_Z the external Zeeman term, H_AF the staggered internal mean-field term (antiferromagnetic) and H_F a small magnetic ordering along the field direction (ferromagnetic). The field-induced ordering is described by the mean-field parameters:

$$H_\mathrm{AF} = -J_t |\langle S^{\perp}_{i,1} \rangle| \sum_j (S^{\perp}_{j,1} - S^{\perp}_{j,2}), \tag{4}$$

$$H_\mathrm{F} = -J_t |\langle S^{z}_{i,1} \rangle| \sum_j (S^{z}_{j,1} + S^{z}_{j,2}), \tag{5}$$

in the notation explained by Lake et al. [9], where J_t are effective couplings. The expectation values of the spin operator $S^{\alpha}_{i,\mu}$ ($\alpha = z$ and $\alpha = \perp$ denote the components parallel and perpendicular to the applied field direction, $\mu = 1, 2$ for the μth ion and $i = 1, \ldots, N$ for the ith dimer) are field-dependent and were taken for H_AF from the elastic neutron-scattering results by Tanaka et al. [5] and for H_F from measurements of the macroscopic magnetization by Shiramura et al. [10]. Qualitative agreement is reported for both observed transitions. Within this model for the effective Hamiltonian a low-energy transition between renormalized $|1, +1\rangle'$ and $|0, 0\rangle'$ states is additionally expected, which was not observed, possibly because of the instrumental resolution or a strong damping. Only at $H = 12\,\mathrm{T}$ there is sevidence for a low-lying mode, which provides a starting point for additional investigations. Conclusive measurements are needed to estimate the accuracy of the model quantitatively and the results will be presented elsewhere [11].

3 Conclusion

The high-energetic spin dynamics in the high-field phase $H > H_\mathrm{c}$ of $TlCuCl_3$ at the minimum of the dispersion can be explained within a conventional Zeeman-type scheme and the effective Hamiltonian in (3). Highly resolved measurements are anticipated, to elucidate the nature of the low-lying signal in the quantum-critical region of the phase diagram. As far as we know, this work represents the first dynamical study of a three-dimensional quantum magnet above the critical field H_c.

Acknowledgements. It is a pleasure to thank M. Meissner and P. Smeibidl for kind support during the measurements on FLEX at HMI (Berlin). Financial support by the Swiss National Science Foundation is gratefully acknowledged.

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