Thermodynamic properties of the field-induced Néel order of TlCuCl$_3$

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Abstract

TlCuCl$_3$ shows a quantum phase transition from a spin-gap phase to a Néel-ordered ground state as a function of magnetic field around $H_{C0} \approx 4.8$ T. From measurements of the specific heat $c$ and the linear thermal expansion $\alpha_i$ we calculate the Grüneisen parameter $\Gamma_i(T) = \alpha_i/c$. Close to $H_{C0}$ we find a diverging $\Gamma_i(T \to 0)$, in qualitative agreement with theoretical predictions. However, the predicted individual temperature dependencies of $\alpha_i(T)$ and $c(T)$ are not reproduced by our experimental data.

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PACS: 75.30.Kz; 75.80.+q; 65.40.De

Keywords: low-dimensional magnets; Bose-Einstein condensation; magnetoelastic coupling; Quantum phase transition

The spin-dimer system TlCuCl$_3$ has been intensively studied in recent years. Besides the intra-dimer coupling $J$ $\approx$ 5.5 meV there are also large inter-dimer couplings $J'$ present [1,2]. The latter cause a strong dispersion of the triplet excitations, and as a consequence the minimum singlet-triplet gap $\Delta_m \approx 0.7$ meV is much smaller than $J$. A moderate magnetic field $H \approx 4.8$ T is already sufficient to close $\Delta_m$ and induces a Néel order with staggered magnetization perpendicular to $H$. In the zero-temperature limit, this transition represents an example of a quantum phase transition [3]. In the vicinity of a quantum critical point (QCP) anomalous temperature dependencies are expected for various physical properties [4]. In particular, a diverging Grüneisen parameter is expected close to a pressure-dependent QCP [5]. Since the phase transition of TlCuCl$_3$ is extremely sensitive to pressure [6,7,8,9] and the control parameter $H$ may be easily tuned, this compound is ideally suited to study such generic properties of a QCP. According to Ref. [5] the following temperature dependencies are expected for $T \to 0$ K close to the QCP of TlCuCl$_3$:

$$c/T \propto \sqrt{T}, \quad \alpha_i/T \propto 1/\sqrt{T}, \quad \text{and} \quad \Gamma_i \propto 1/T.$$  \hspace{1cm} (1)

Here, $c$ denotes the specific heat, $\alpha_i$ is the linear thermal expansion coefficient along the direction $i$ and $\Gamma_i = \alpha_i/c$ the respective Grüneisen parameter.

We present high-resolution measurements of the uniaxial thermal expansion $\alpha_i = 1/L_i \cdot \partial L_i/\partial T$ along different lattice directions $i$ ($L_i$ is the respective sample length along $i$) and the specific heat $c$ for $T \gtrsim 0.5$ K. Since TlCuCl$_3$ easily cleaves along the (010) and (10¯) planes of the monoclinic structure, we measured $L_i(H,T)$ perpendicular to these planes on a single crystal of dimensions $1.7 \times 1.5 \text{ mm}^2$ perpendicular to (010) and (10¯), respectively. In addition, the (201) direction was measured on a second crystal of length $L_{[201]} = 4.4 \text{ mm}$. All properties have been studied up to $H = 14$ T applied perpendicular to the (102) plane.

Fig. 1 shows $\alpha_i$ measured along all three directions for different magnetic fields. In zero field, $\alpha_i/T$ continuously approaches zero for $T \to 0$. With increasing $H$ a pronounced shoulder develops, which reaches a maximum around 4.5 T. For larger fields clear anomalies with a sign change of $\alpha_i/T$ occur. These anomalies systematically sharpen and shift towards higher $T$ with further increasing field. From magnetostriiction measurements (not shown) we find a critical field $H_{C0} \approx 4.8$ T for $T \to 0$. The broad anomalies appear in the $\alpha_i(T)$ curves already for $H \gtrsim 4.6$ T. We attribute this to the finite width of the phase transition. The curves for $i = (010)$ and (102) agree well with our previous results measured on a different crystal for $T \gtrsim 3 \text{ K}$ [8,10]. There we have also shown that the $\alpha_i$ anomalies signal huge uniaxial pressure dependencies of $T_N$, which arise from the pressure-dependent changes of the intradimer coupling $J$. From the sum of all three $\alpha_i$ we can...
also determine the volume changes and calculate the hydrostatic pressure dependence of the spin gap. We obtain $\partial \ln \Delta_m / \partial p_{\text{hydro}} \simeq -300 \% / \text{GPa}$, in reasonable agreement with the initial slope of $\simeq -400 \% / \text{GPa}$ found by magnetization measurements under hydrostatic pressure [9].

In Fig. 2 we show the specific heat for different fields. In agreement with Ref. [11] we find a rather small anomaly even for the largest field, and the magnitude rapidly decreases when $H_{C0}$ is approached. From $\alpha_i$ and $c$ we calculated the uniaxial Grüneisen parameters $\Gamma_i$ shown in Fig. 3. For all three directions $\Gamma_i$ remains finite for $T \to 0$ in zero and small fields. However, all three $\Gamma_i$ show a clear tendency to diverge for $H \simeq 4.5 \text{ T}$. In the upper right panel of Fig. 3 we display $\Gamma_i(T)$ for $H \lesssim H_{C0}$ on double-logarithmic scales. Within experimental accuracy, the slope of all three $\Gamma_i(T)$ is identical. A power-law fit yields $\Gamma_{(010)} \propto T^{-4/3}$ and describes the experimental data reasonably well for about one decade. Since only the irregular contributions of $c$ and $\alpha_i$ are considered in Ref. [5], while the experimental data also contain the respective phononic contributions, one may tend to the conclusion that our data nicely confirm the theoretical prediction $\Gamma(T) \propto T^{-1}$. However, the agreement between theory and experiment is much worse when the individual temperature dependencies of $c/T$ and $\alpha_i/T$ are considered. Neither the data of Fig. 1 nor those of Fig. 2 give any indication to follow the predicted temperature dependencies of Eq. (1). Concerning the specific heat data, one may argue that the predicted $\sqrt{T}$ behavior is difficult to identify because of the phononic contribution. However, this argument does not hold for $\alpha_i$, since the predicted divergence of $\alpha_i/T$ should be seen despite a (regular) phononic contribution. Thus, our experimental data do only partially confirm the theoretical predictions [5].

In summary, we find diverging Grüneisen parameters $\Gamma_i = \alpha_i/c$ close to the quantum critical point of TlCuCl$_3$, in qualitative agreement with theoretical predictions. However, $\alpha_i$ and $c$ do not show the predicted temperature dependencies. One may suspect that this disagreement could arise from the magnetic anisotropy of TlCuCl$_3$, which is not considered in Ref. [5], or from a broadening of the transition due to sample imperfections, as e.g. internal strains.

We acknowledge discussions with A. Rosch, I. Fischer, and J.A. Mydosh. This work was supported by the DFG via SFB 608.

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