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Field induced magnetic phase transition as a magnon Bose Einstein condensation

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Abstract

We report specific heat, magnetocaloric effect and magnetization measurements on single crystals of the frustrated quasi-2D spin $-\frac{1}{2}$ antiferromagnet Cs_2CuCl_4 in the external magnetic field $0 \leqslant B \leqslant 12\,T$ along a-axis and in the temperature range $0.03\,K \leqslant T \leqslant 6\,K$. Decreasing the applied magnetic field B from high fields leads to the closure of the field induced gap in the magnon spectrum at a critical field $B_c \simeq 8.44\,T$ and a long-range incommensurate state below B_c. In the vicinity of B_c, the phase transition boundary is well described by the power law $T_N \sim (B_c - B)^{1/\phi}$ with the measured critical exponent $\phi \simeq 1.5$. These findings provide experimental evidence that the scaling law of the transition temperature T_N can be described by the universality class of 3D Bose–Einstein condensation (BEC) of magnons.

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Keywords: Specific heat; Quantum critical point; Bose–Einstein condensation

1. Introduction

Cs_2CuCl_4 was proposed recently as a quasi-two-dimensional (2D) Heisenberg antiferromagnet (AFM) with $S = \frac{1}{2}$ Cu^{2+} spins arranged in triangular lattices (bc plane) which are weakly coupled along the crystallographic a direction [1]. The weak interlayer couplings stabilize magnetic order at temperatures below 0.62 K. In the ordered state, magnetic moments form an incommensurate spiral in the (b, c) plane. The system enters a fully spin-polarized state at high magnetic field B exceeding the saturation field $B_c \simeq 8.5\,T$ [2–4]. In this state spin excitations are gapped ferromagnetic (FM) magnons and have a quadratic energy dispersion near the magnon band minima. In the opposite direction, i.e., with decreasing B and passing through B_c, the field induced magnon gap disappears at B_c and an AFM long range-order with the transverse spin component develops [1–3]. In this system the dominant exchange spin coupling (along b) J is rather weak, $J = 4.34(6)\,K$ [1]. There are also isotropic exchange couplings along the zig-zag bonds in the (b, c) plane, $J' = 0.34(3)J$ and moreover the anisotropic Dzyaloshinsky–Moriya (DM) interaction [5] determined with high accuracy by neutron experiments [2]. Thus, the spin Hamiltonian involves the isotropic exchange H_0, the DM anisotropic term H_{DM}, and the Zeeman energy H_B. The DM interaction breaks the isotropic spin symmetry and creates an easy-plane anisotropy in the (b, c) plane of the spiral order. Thus, Cs_2CuCl_4 falls into the class of easy-plane AFMs with $U(1)$-rotational invariance around the crystallographic a-axis.

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Thus, for B applied along the a-axis, the $U(1)$symmetry can be broken spontaneously due to the transverse spin component ordering at T_c. However, unambiguous evidence for a Bose–Einstein condensation (BEC) description of the field-induced phase transition would be the determination of the critical exponent ϕ in the field dependence of the critical temperature

$$T_c(B) \propto (B_c - B)^{1/\phi}. \quad (1)$$

Theory for a 3D Bose gas predicts a universal value $\phi_{BEC} = \frac{3}{2}$ [6,7], which coincides with the result of a mean-field treatment [8].

A magnon BEC in $TlCuCl_3$ was recently reported [8,9]. In this quantum AFM with a dimerized spin–liquid ground state, the saturation field is rather high, $B_{c,2} \approx 60$ T, and the BEC transition was studied near the first critical field, $B_{c,1} \simeq 5.6$ T. At $B = B_{c,1}$, the singlet-triplet excitation gap is expected to close and a BEC occurs for $B > B_{c,1}$ [8]. However, a few experimental findings show deviations from a pure magnon BEC: An anisotropic spin coupling (of unknown nature) might produce a small but finite spin gap in the ordered state for $B > B_{c,1}$ [10], and the reported critical exponent ϕ is somewhat larger than the value predicted by theory [8]. Although there are a number of other excellent realizations of similar quantum systems, Cs_2CuCl_4 is one of the few quasi-2D systems with a relatively weak (~4 K) exchange interactions which can be overcome by current fields (9 T). Moreover, the access to very low temperatures ($T/J \simeq 10^{-2}$) enabled us to be as close as possible to the asymptotic regime where universal scaling laws are expected to hold.

2. Experiment

In this work, we report on specific heat, magnetocaloric effect, and magnetization measurements on high quality single crystal of Cs_2CuCl_4 at low temperatures ($0.03\,\mathrm{mK} < T < 6$ K) and magnetic fields applied along the crystallographic a-axis. The specific heat measurements were performed at temperatures down to 0.03 K and magnetic fields up to 11.5 T using the compensated quasiadiabatic heat pulse method [11]. Using the same experimental setup, the magnetocaloric effect in Cs_2CuCl_4 was studied in detail by measuring the temperature of the sample in a quasiadiabatic regime as a function of magnetic field. The magnetization was measured at temperatures down to 0.05 K and high fields up to 11.5 T using a high resolution capacitive Faraday magnetometer [4]. The aim of this study was: (i) to trace the field dependence of $T_c(B)$ near B_c, i.e., to extract the power law according to Eq. (1), and (ii) to observe the closure of the spin gap.

3. Results and discussion

The specific heat of Cs_2CuCl_4 in zero magnetic field and for fields $B \leqslant B_c$ is shown in Fig. 1. The magnetic contribution C_{mag} to the total specific heat C_{tot} was

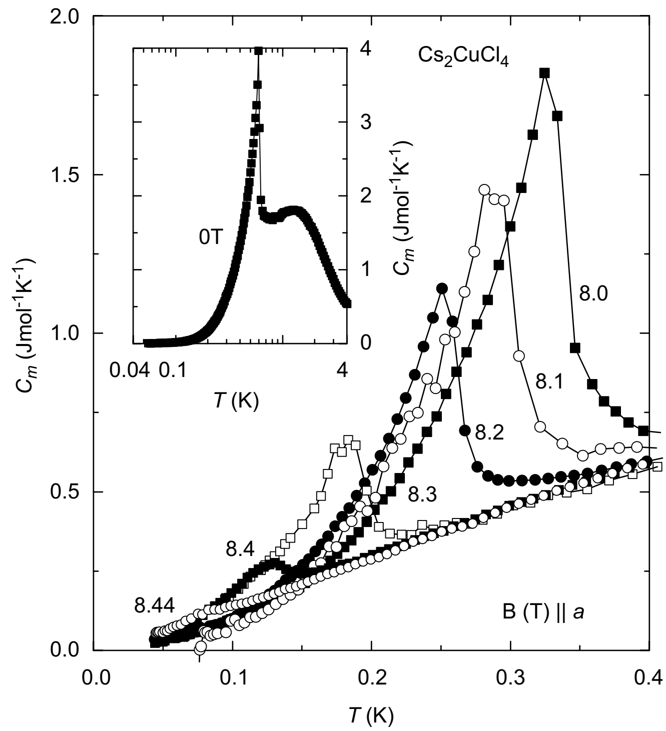

Fig. 1. Temperature dependence of the magnetic specific heat C_{mag} of Cs_2CuCl_4 close to the critical field. Inset: $C_{mag}(T)$ for zero magnetic field.

obtained by subtracting the phonon contribution $C_{ph} = 1.36 \times 10^4 (T/\Theta_D)^3\,\mathrm{J\,mol^{-1}\,K^{-1}}$ (using a Debye temperature $\Theta_D = 126$ K) and the nuclear contribution $C_{nuc} = \alpha/T^2$. The most prominent features of the magnetic specific heat at $B = 0$, presented in Fig. 1 [1] are a sharp λ-like anomaly at $T_N = 0.62$ K which indicates the transition to 3D long-range order, where the magnetic structure found by neutron diffraction is a spiral in the (b, c) plane [12], and a broad maximum of height $C/\mathrm{R} = 0.35 \pm 0.01$ at $T_{max} \simeq 2$ K. The positions of ordering temperature and the broad maximum change only slightly for small fields. However, the transition temperature to the spiral ordered state, which can be regarded as a conelike structure in field [2], varies very strongly above 8 T (Fig. 1). The λ-like anomaly in $C_{mag}(T)$ is gradually suppressed in its height, and its position is pushed to lower temperatures with increasing field. As the field is increased from 8.4 to 8.44 T (Fig. 1) T_c is reduced by almost a factor of 2 ($T_c = 76$ mK at $B = 8.44$ T), and T_c has shifted downwards by almost 1 order of magnitude compared to the zero-field value. No further signatures of the transition can be resolved in our data at higher fields. Complementary magnetization measurements were performed on Cs_2CuCl_4. Susceptibility $\chi \simeq M/B$ in a field of 0.1 T is shown in Fig. 2. It displays a Curie–Weiss local-moment behavior at high temperatures and a broad maximum around $T_{max} = 2.81(1)$ K, characteristic of short-range antiferromagnetic correlations. No clear anomaly at T_N is observed and this is because the magnetic structure has ordered moments spiralling in a plane which makes a very small angle (~17°) with the b, c

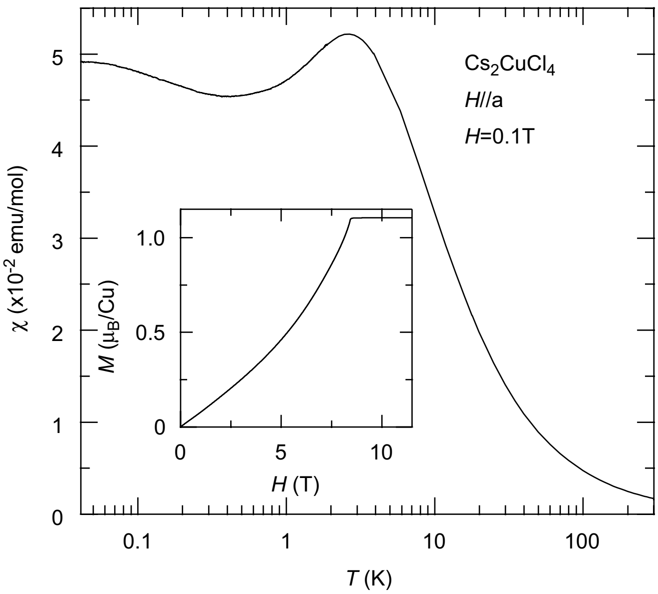

Fig. 2. Temperature dependence of the susceptibility $\chi = M/B$ of Cs_2CuCl_4 along the crystallographic *a*-axis. Inset: Magnetization curve of Cs_2CuCl_4 measured at the base temperature for the field applied along the crystallographic *a*-axis.

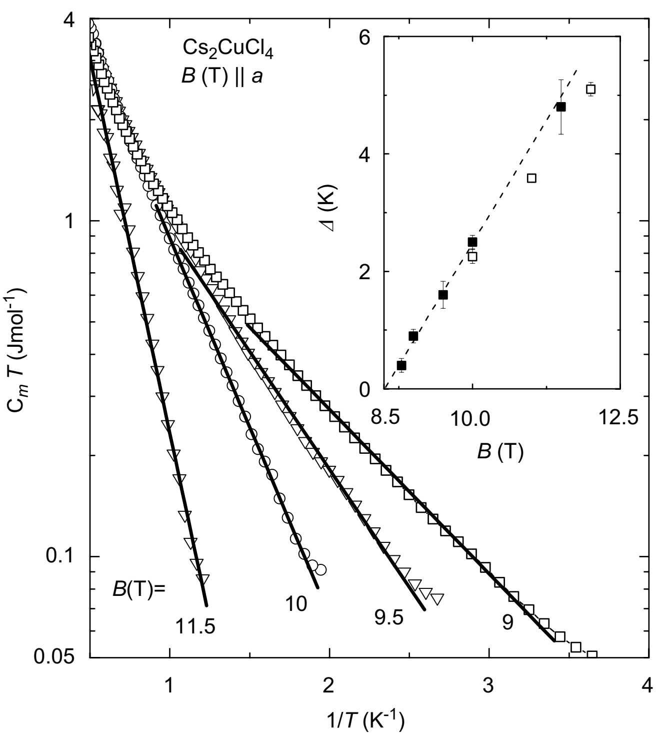

Fig. 3. Semilogarithmic plot of $C_{mag}T$ vs $1/T$ of Cs_2CuCl_4 for fields above B_c. The slope of the data (solid lines) yields the value of the gap Δ present in the magnon excitation spectrum. Inset: $\Delta(B)$ extracted from $C(T)$ (black symbols) and neutron scattering measurements respectively (white symbols). The dashed line represents a linear fit to the data which yields B_c for $\Delta = 0$.

plane [12]. In this case the near out-of-plane (*a*-axis) susceptibility is much less sensitive to the onset of magnetic order compared to the in-plane susceptibility (along *b* and *c*) [4]. Inset of Fig. 2 shows the magnetization $M(B)$ of Cs_2CuCl_4 as a function of the applied magnetic field at 0.05 K. At low fields, the magnetization increases linearly but has a clear overall convex shape. It saturates above the critical field $B_{sat} = 8.44(2)$ K when the system enters the field induced FM state in good agreement with the above specific heat measurements. In the FM state, neutron scattering measurements have revealed a field-induced gap in the magnon excitation spectrum [2]. For the interpretation of the phase transition below B_c as a BEC of magnons it is crucial that the gap closes at B_c. To provide compelling evidence, we analyzed the $C(T)$ data above B_c. Assuming first a 2D quadratic magnon dispersion, the leading contribution to the temperature dependence of the specific heat is given by $C_{mag} \simeq \exp(-\Delta/T)/T$, provided that $T < \Delta$. This behavior fits well the experimental data at low temperatures (see Fig. 3).

The determined values for the gap are shown in the inset of Fig. 3. The dashed line represents a linear fit to the data which yields B_c for $\Delta \to 0$.

However, for a more thorough test of the predicted power-law field dependence of the critical temperature $T_c(B) \sim (B_c - B)^{2/3}$ it is important to perform the fitting procedure in a narrow field window close to B_c where the theoretical predictions are valid. Since B_c is almost temperature independent at low temperatures, a measurement with temperature sweep detects broad anomaly at T_c while a field sweep measurement can probe more precisely the transition points in (T, B) phase diagram. Therefore, in addition to specific heat data, we also measured the

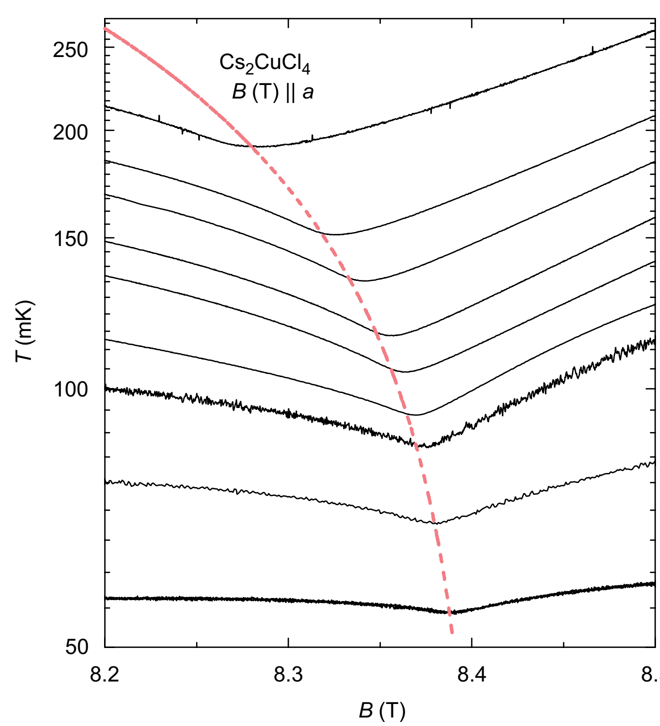

Fig. 4. Temperature change upon adiabatic field scans. Dashed line represents a best fit to a power law form for the phase boundary with exponent $\phi = 1.55(5)$ as described in the text.

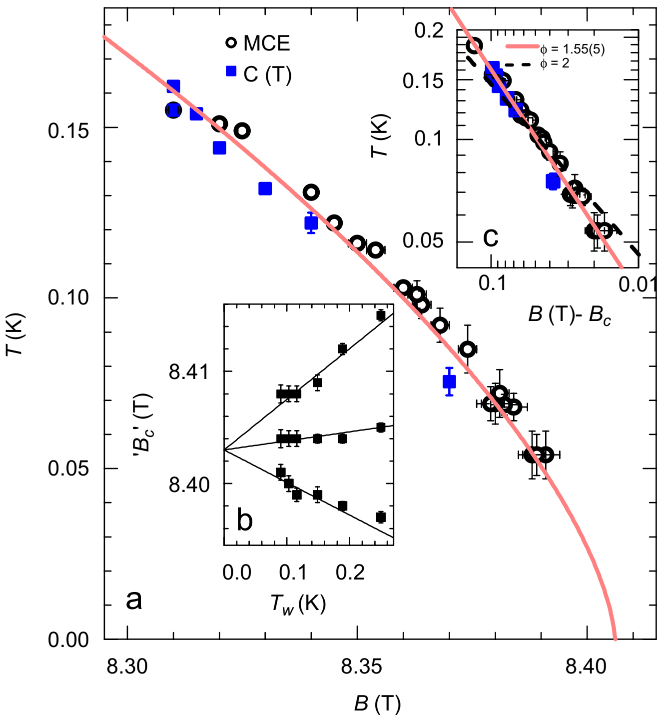

Fig. 5. $T_N(B)$ data on linear (a) and log scales (b). The solid line in (a) is a power-law fit with $\phi = 1.55(5)$; the dashed line in (b) represents a power law curve for $\phi = 2$. (b) Estimates of the critical field B_c obtained from power-law fits to the low temperature $T \leqslant T_w$ data at fixed $\phi = 1.6, 1.5$, and 1.4 (top to bottom).

magnetocaloric effect (MCE) to follow the suppression of magnetic order by the applied field down to very low temperatures (50 mK). The phase boundary between the low-field cone ordered phase and the paramagnetic phase based on MCE is shown in Fig. 5(a). The data determined from locations of sharp peaks in $C(T)$ and minima in the fields scans of the MCE are shown in Fig. 4. The MCE effect describes the variation of the sample temperature upon adiabatically varying the field and anomalies occur near the phase transitions. For a second-order phase transition line that ends in a $T = 0$ quantum critical point as it is the case here, the change of sign of $\Gamma_B = T^{-1}(\mathrm{d}T/\mathrm{d}B)_S$ at sufficiently low T occurs very close to the actual phase boundary $T_c(B)$ [13]; the observed overlap between the specific heat points of $T_c(B)$ and location of the MCE anomaly already at 0.15 K (see Fig. 5(a)) shows that this criterion is well satisfied here.

Since a two parameter fit of the phase boundary data in Eq. (1) with both B_c and exponent ϕ varying can still be questioned [7,14], we applied a procedure proposed in [14] for an independent determination of B_c. The power-law given above was fitted to the lowest-temperature data points in a temperature window $T \leqslant T_w$ of gradually increasing size for several fixed exponents ϕ. The obtained critical field values B_c are plotted in Fig. 5(b) as a function of T_w and their linear extrapolation to $T_w = 0$ shows good convergence to $B_c = 8.403(4)$ T (solid lines in Fig. 5(c)). This value for the critical field was then used in the power-law fit to the data below 0.17 K (over 20 points) and gave the exponent $\phi = 1.55 \pm 0.05$ (solid line in Fig. 5(a) and (b)), in good agreement with the BEC prediction of $\phi = 1.5$.

4. Conclusions

In conclusion, using thermodynamic and magnetic measurements, we determined the phase diagram (T, B) of Cs_2CuCl_4 along the crystallographic a-axis in detail. The observed scaling of the critical temperature close to the saturation field is in good agreement with predictions of 3D BEC in a dilute gas of magnons. Together with the observed opening of a spin gap above B_c these findings support the notion of a BEC of magnons in Cs_2CuCl_4.

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