Bose-Einstein Condensation of Magnons in Cs_2CuCl_4

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In a quantum antiferromagnet (AFM) a fully spin-polarized state can be reached at high magnetic field B exceeding a saturation field B_c. In this state, spin excitations are gapped *ferromagnetic* magnons. With decreasing B and passing through B_c, an antiferromagnetic long-range order of the transverse spin component develops. Provided the symmetry of the spin Hamiltonian is such that the rotational invariance around the applied field is preserved, the transverse spin component ordering can be regarded as a Bose-Einstein condensation (BEC) in a dilute gas of *antiferromagnetic* magnons [1,2]. For most of the known AFMs, B_c can be well above 100 T. An exceptionally low and easily accessible saturation field of $B_c \approx 8.5$ T is, however, needed in the quantum spin-1/2 AFM Cs_2CuCl_4. In this system the dominant exchange spin coupling J is rather weak, $J = 4.34(6)$ K [3]. The other isotropic spin coupling constants and the anisotropic Dzyaloshinsky-Moriya (DM) interaction are smaller and were determined with high accuracy by neutron scattering experiments [3]. Thus, the spin Hamiltonian involves the isotropic exchange H_0, the DM anisotropic term H_{DM} and the Zeeman energy H_B.

Cs_2CuCl_4 falls into the class of easy-plane AFMs with $U(1)$-rotational invariance around the crystallographic a-axis. 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. This is accompanied by the appearance of a Goldstone mode with linear dispersion, which is interpreted as signature of a magnon BEC [3]. However, an unambiguous evidence for a 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

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

Theory for a 3D Bose gas predicts a universal value $\phi_{BEC} = 3/2$ [4], which coincides with the result of a mean-field treatment [5].

We performed specific-heat measurements on single crystals of Cs_2CuCl_4 at low temperatures ($30\text{ mK} < T < 6$ K) and magnetic fields applied along the crystallographic a-axis [6]. Figure 1 shows the magnetic contribution, $C_{mag}(T)$, to the total specific heat of Cs_2CuCl_4 at fields close to B_c. 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 upon approaching the critical field $B_c \simeq 8.5$ T. For $B > B_c$ the ordering of the transverse component of the magnetic moment completely disappears since the spin system enters a field-induced ferromagnetic (FM) state [3]. For the interpretation of the phase transition below B_c as a BEC of magnons it is crucial that the gap in the ferromagnetic magnon excitation spectrum present above B_c closes as B_c is approached from high fields. The compelling evidence for this is presented in the inset to Fig. 1. Assuming 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 $T < \Delta$. As shown in the inset to Fig. 1, this behavior fits well the experimental data above 0.3 K.

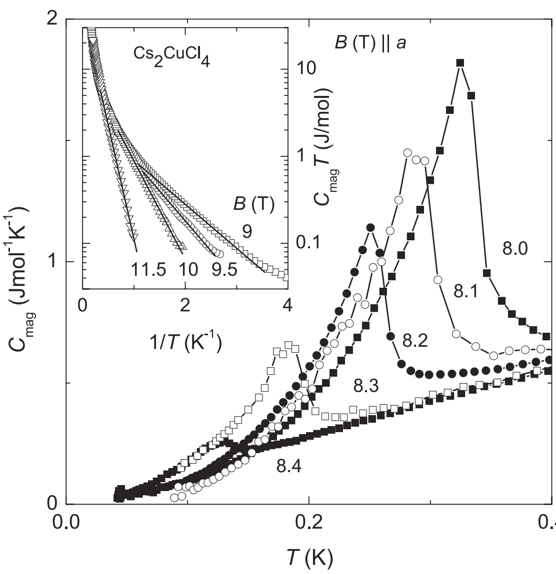

Fig. 1: Magnetic specific heat, C_{mag} vs. T of Cs_2CuCl_4 close to the critical field. Inset: Semi-logarithmic plot of $C_{mag}T$ vs. $1/T$ of Cs_2CuCl_4 for fields above B_c. In this representation the slope of the data (solid lines) yields the value of the gap Δ present in the magnon excitation spectrum.

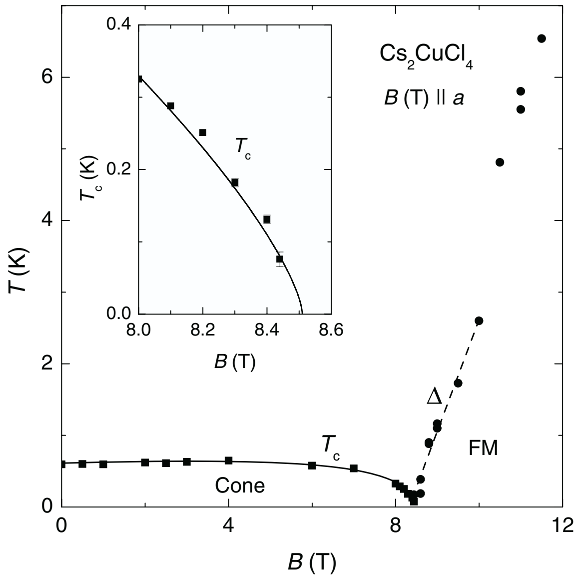

Fig. 2: (T, B) phase diagram of Cs_2CuCl_4 for $B \parallel a$. Upon approaching the critical field B_c = 8.51 T the ordering temperature T_c approaches 0 K. Above B_c the gap Δ in the spin excitation spectrum opens. Inset: The experimental $T_c(B)$ data points and the calculated phase boundary of the BEC of magnons (solid line) are in very good agreement.

This analysis leads to the (T, B) phase diagram of Cs_2CuCl_4 presented in Fig. 2. T_c starts to decrease strongly above 8 T and $T_c \rightarrow 0$ for $B \rightarrow B_c$. Fitting the power law dependence $T_c(B) \propto (B_c - B)^{1/\phi}$ to the data for $B \geq 8$ T with the assumption of $B_c = 8.50$ T yields an exponent $\phi = 1.52(10)$. The linear extrapolation $\Delta \rightarrow 0$ of the field dependence of the gap yields $B_c = 8.3(10)$ T and $g = 2.31(15)$. The relatively large errors are due to the uncertainties in the fit. It is noteworthy that the experimental data and the solid line plotted in the inset to Fig. 2 are in excellent agreement. Our theoretical analysis of the data is described below.

To treat the observed phase transition slightly below the saturation field B_c as a BEC of magnons [1,2,5] we used the hard-core boson representation for spin-1/2 operators $S_i^{\pm}$, S_i^z in the original Hamiltonian H. Due to the sign change of the DM interaction ($D = 0.053(5)\,J$ [3]) between even and odd magnetic layers, which are stacked along the a-direction, two types of bosons, a_i and b_j are introduced for the two types of layers [7]. The hard-core boson constraint was satisfied by adding to H an infinite on-site repulsion, $U \rightarrow \infty$, between bosons:

$$H_U^{(a)} + H_U^{(b)} = U \sum_i a_i^+ a_i^+ a_i a_i + U \sum_j b_j^+ b_j^+ b_j b_j . \quad (2)$$

The interlayer coupling $J'' = 0.045(5)\,J$ [3] mixes a and b boson modes and results in two bare magnon excitation branches A and B. Their dispersion relations are [3]

$$E_q^{A,B} = J_q \mp \operatorname{sign} D_q \sqrt{D_q^2 + \left(J_q''\right)^2} - E_0, \quad (3)$$

with

$$J_q = J \cos q_x + 2J' \cos(q_x/2) \cos(q_y/2) \ , \quad (4)$$
$$D_q = 2D \sin(q_x/2) \cos(q_y/2) \ , \quad (5)$$
$$J_q'' = J'' \cos(q_z/2) \ . \quad (6)$$

Here $J' = 0.34(3)\,J$ [3] and the q-values are restricted to $0 \leq q_x < 2\pi$, $0 \leq q_y < 4\pi$, and $0 \leq q_z < 2\pi$.

The degenerate minima $E^A_{Q_1} = E^B_{Q_2} = 0$ are at $Q_1 = (\pi + \delta_1, 0, 0)$ for branch A and $Q_1 = (\pi - \delta_2, 2\pi, 0)$ for branch B. Without loosing precision we can use $\delta_1 \simeq \delta_2 \simeq 2 \arcsin(J'/2J)$. Then the bilinear part of H is

$$H_{bil} = \sum_q \left[\left(E_q^A - \mu_0\right) A_q^+ A_q + \left(E_q^B - \mu_0\right) B_q^+ B_q \right], \quad (7)$$

with $A_q = \alpha_q a_q + \beta_q b_q$, $B_q = \alpha_q b_q + \beta_q a_q$, $\alpha_q^2 + \beta_q^2 = 1$, and $\mu_0 = g\mu_B(B_c - B)$ the bare chemical potential. $B_c = W/(g\mu_B)$, with W being the magnon bandwidth, was calculated to be $B_c = 8.51$ T assuming $g = 2.20$ [7].

The interaction given by eq. 2 describes the scattering of A and B magnons. Near the quantum critical point, $(B_c - B) \ll B_c$ and at low temperature, the average density of magnons $n^A = n^B = n$ is low, $n \sim (1 - B/B_c)$. The magnon scattering can be treated in the ladder approximation, neglecting interference between a and b channels. In this approximation, the problem reduces to solving the Bethe-Salpeter equation in each channel.

For a given $B \lesssim B_c$ and with decreasing temperature the magnon BEC occurs when the effective chemical potential μ_{eff} vanishes [5]. Then $T_c(B)$ is determined by

$$g\mu_B(B_c - B) = 2\Gamma n(T_c). \quad (8)$$

Here $n(T) = \Sigma_q f_B(E_q)$ with $f_B(E_q)$ being the Bose distribution function taken at $\mu_{\text{eff}} = 0$ and $E_q = E_q^A$ or $E_q = E_q^B$. This means that for $T < T_c$ the magnon condensate develops simultaneously at $q = Q_{1,2}$. It is worth emphasizing that at $\mu_{\text{eff}} \rightarrow 0$ the distribution function $f_B(E)$ diverges as T/E for $E \rightarrow 0$. Therefore, the low energy 3D-magnon spectrum, $E < E^*$, mainly contributes and drives the BEC transition. The phase boundary can be calculated using eq. 8. It gives a very good description of the experimental data near B_c (see inset to Fig. 2), but deviates strongly at lower fields, i.e., for $B_c – B > 0.5$ T. This indicates that the mean-field description of the magnon BEC is only applicable in the close vicinity of B_c. The calculated boundary is well described by eq. 1 with a critical exponent $\phi_{th} \simeq 1.5$ close to the predicted value $\phi_{\mathrm{BEC}} = 3/2$ characteristic for 3D quadratic dispersion of low-energymagnons.

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