Bose-Einstein Condensation of Magnons in Cs$_2$CuCl$_4$

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We report on results of specific heat measurements on single crystals of the frustrated quasi-2D spin-1/2 antiferromagnet Cs$_2$CuCl$_4$ ($T_N = 0.595$ K) in external magnetic fields $B < 12$ T and for temperatures $T > 30$ mK. Decreasing $B$ from high fields leads to the closure of the field-induced gap in the magnon spectrum at a critical field $B_c \approx 8.51$ T and a magnetic phase transition is clearly seen below $B_c$. In the vicinity to $B_c$, the phase transition boundary is well described by the power-law $T_c(B) \propto (B_c - B)^{1/\phi}$ with the measured critical exponent $\phi \approx 1.5$. These findings are interpreted as a Bose-Einstein condensation of magnons.

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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 magnons. This concept was formulated theoretically many years ago $\textbf{[1,2]}$. 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, however, is needed in the quantum spin-1/2 AFM Cs$_2$CuCl$_4$. In this system the dominant exchange spin coupling $J$ is rather weak, $J = 4.34(6)$ K $\textbf{[3]}$. The other isotropic spin coupling constants and the anisotropic Dzyaloshinsky-Moriya (DM) interaction are smaller and were determined with high accuracy by neutron experiments $\textbf{[1]}$. Thus, the spin Hamiltonian involves the isotropic exchange $H_0$, the DM anisotropic term $H_{DM}$ and the Zeeman energy $H_B$ and is given by $H = H_0 + H_{DM} + H_B$.

Cs$_2$CuCl$_4$ 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 $\textbf{[4]}$. However, an unambiguous evidence for a BEC description of the field-induced phase transition would be the determination of the critical exponent $\phi$ in the field dependence of the critical temperature

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

Theory for a 3D Bose gas predicts a universal value $\phi_{\text{BEC}} = 3/2$ $\textbf{[5]}$, which coincides with the result of a mean-field treatment $\textbf{[6]}$.

A magnon BEC in TICuCl$_3$ was recently reported $\textbf{[6,7]}$. 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, $B_{c,1} \approx 5.6$ T. At $B = B_{c,1}$, the singlet-triplet excitation gap is expected to close and a BEC occurs for $B > B_{c,1}$ $\textbf{[3]}$. 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 $B > B_{c,1}$ $\textbf{[8]}$ and the reported critical exponent $\phi$ is somewhat larger than predicted by theory $\textbf{[3]}$.

In this Letter we report on specific heat measurements $\textbf{[9]}$ on single crystals of Cs$_2$CuCl$_4$ at low temperatures ($30$ mK $< T < 6$ K) and high magnetic fields ($B < 12$ T) applied along the crystallographic $a$-axis. The aim of this thermodynamic study was (i) to trace the field dependence of $T_c(B)$ near $B_{c,1}$, i.e., to extract the power law according to eq. $\textbf{1}$ and (ii) to determine the closure of the spin gap. The access to very low temperatures ($T/J \approx 10^{-2}$) enabled us to be as close as possible to the asymptotic regime where universal scaling laws are expected to hold.

Figure $\textbf{[10]}$ shows the specific heat of Cs$_2$CuCl$_4$ in zero magnetic field. The magnetic contribution, $C_{mag}$ to the total specific heat, $C_{tot}$, was obtained by subtracting the phonon contribution $C_{ph} = 13599 (T/\Theta_D)^3$ Jmol$^{-1}$K$^{-1}$ (using a Debye temperature $\Theta_D = 126$ K). Furthermore, the contribution of the nuclear specific heat to the total specific heat at very low temperatures has been accounted for in the analysis of all the raw data shown in the following $\textbf{[10,11]}$. The two prominent features present in $C(T)$ are the broad maximum related to the cross-over from the paramagnetic to a short-range spin correlated state and the $\lambda$-like peak. The latter is the signature of the entrance into the 3D magnetically ordered state ($T_N = 0.595$ K), where the magnetic structure is a spiral in the $(b,c)$ plane $\textbf{[3]}$. The overall shape of $C(T)$ above $T_N$ is already captured quantitatively by including only the strongest term in the Hamiltonian, including only the strongest term in the Hamiltonian, including only the strongest term in the Hamiltonian,
The ordering temperature and the position of the broad maximum hardly change for small fields. However, the transition temperature to the spiral ordered state, which can be regarded as a cone-like structure, varies very strongly above 8 T (Fig. 2). Instead of $T_N$, we label the transition temperature in this field range $T_c$, as in eq. 1 in order to follow the nomenclature used in the theoretical description. The $\lambda$-like anomaly in $C_{\text{mag}}(T)$ is gradually suppressed in its height and its position is pushed to lower temperatures with increasing field. An extraordinary change occurs as the field is increased from 8.4 T to 8.44 T (inset to Fig. 2). Upon this tiny field change ($\Delta B/B < 0.5\%$), $T_c$ is reduced by almost a factor of two ($T_c = 76$ mK at $B = 8.44$ T), and $T_c$ has shifted downwards by almost one order of magnitude compared to the zero-field value. No further signatures of the transition can be resolved in our data upon approaching the critical field $B_c \approx 8.5$ 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.

A field-induced gap in the magnon excitation spectrum was observed in the FM state by neutron scattering measurements. Its field dependence was given to be $\Delta = g\mu_B(B - B_c)$, with $B_c \approx 8.44$ T, $g = 2.18$, and $\mu_B$ the Bohr magneton. 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 a compelling evidence for this fact we re-examined the phase diagram above $B_c$ with our thermodynamic measurements. The results are presented in Fig. 3.

The magnon dispersion along the $a$-direction is small due to a weak interlayer spin coupling. Thus, for temperatures well above a characteristic energy scale $E^* \approx 50$ mK, the actual magnon dispersion is of 2D character. However, for $T < E^*$ a smooth cross-over to a 3D character is expected. Assuming first a 2D quadratic magnon dispersion, the leading contribution to the temperature dependence of the specific heat is given by $C_{\text{mag}} \approx \exp(-\Delta(T)/T)$, provided that $T < \Delta$. As shown in Fig. 3, this behavior fits well the experimental data above 0.3 K. The obtained field dependence of $\Delta(B)$ is discussed below. The deviation from a straight line of the 9 T data below 0.3 K in Fig. 3 might indicate the cross-over from 2D to 3D magnons. This notion is supported by the low-temperature data plotted as $C_{\text{mag}}\sqrt{T}$ vs $1/T$ in the inset to Fig. 3. This presentation is used
since a 3D dispersion relation yields as leading term in the specific heat \( C_{\text{mag}} \approx \exp(-\Delta/T)/\sqrt{T} \). The straight line below \( \approx 0.15 \) K indicates that this model describes the data equally well. However, in this cross-over region, the 2D model describes the data equally well and the determined value of the gap \( \Delta \) is (within the error bar) the same as the one deduced from the \( C_{\text{mag}}T \) vs \( 1/T \) plot.

The \((T, B)\) phase diagram of Cs\(_2\)CuCl\(_4\) obtained from our specific heat experiments is presented in Fig. 4. The field dependence of the magnetic transition temperature is in very good agreement with \( T_c(B) \) obtained from neutron data up to 8 T [14]. The specific heat data, however, revealed that \( T_c \) starts to decrease strongly above 8 T and \( T_c \to 0 \) for \( B \to B_c \). 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 \( \phi = 1.52(10) \). We want to stress that the value of \( \phi \) is very sensitive to the chosen value of \( B_c \). An exponent \( \phi = 1.44(10) \) is obtained if \( B_c = 8.51 \) is used. The solid line plotted in the inset to Fig. 4 represents the result of the theoretical analysis described below. Above \( B_c \) the fully spin-polarized FM state is created and the gap \( \Delta \) opens in the spin excitation spectrum. The dashed line represents a linear fit to the data for \( B \leq 10 \) T (main part of Fig. 4). This yields \( B_c = 8.3(10) \) T and \( g = 2.31(15) \). The relatively large errors are due to the uncertainties in the fit.

To treat the observed phase transition slightly below the saturation field \( B_c \) as a BEC of magnons [1, 2, 6], we used the hard-core boson representation for spin-1/2 operators \( S_i^x, S_i^y \) in the original Hamiltonian \( H \). Because the DM interaction \((D = 0.053(5)J) \) changes sign 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 [15, 16]. The hard-core boson constraint was satisfied by adding to \( H \) an infinite on-site repulsion, \( U \to \infty \), between bosons given by

\[
H_{U}^{(a)} + H_{U}^{(b)} = U \sum_i a_i^+ a_i + U \sum_j b_j^+ b_j.
\] (2)

The interlayer coupling \( J'' = 0.045(5)J \) mixes \( a \) and \( b \) boson modes and results in two bare magnon excitation branches \( A \) and \( B \). Their dispersion relations are given by [1]

\[
E_q^{A,B} = J_q \mp \text{sign} D_q \sqrt{D_q^2 + (J''_q)^2} - E_0,
\] (3)

with

\[
J_q = J \cos q_x + 2J' \cos (q_x/2) \cos (q_y/2),
\] (4)

\[
D_q = 2D \sin (q_x/2) \cos (q_y/2),
\] (5)

\[
J''_q = J'' \cos (q_z/2).
\] (6)
Here \( J' = 0.34(3)J \) [1] and the \( q \)-values are restricted to 
\( 0 \leq q_x < 2\pi, \ 0 \leq q_y < 4\pi, \ \text{and} \ 0 \leq q_z < 2\pi. \)

The degenerate minima \( E^A_{\vec{Q}_1} = E^B_{\vec{Q}_2} = 0 \) are at \( \vec{Q}_1 = (\pi + \delta_1, 0, 0) \) for branch \( A \) and at \( \vec{Q}_2 = (\pi - \delta_2, 2\pi, 0) \) for branch \( B. \) Without loosing precision we can use \( \delta_1 \approx \delta_2 \approx \frac{\pi}{2} \text{arcsin}(J'/2J). \) The bilinear part of \( H \) now reads

\[
H_{\text{bil}} = \sum_q \left[ (E^A_q - \mu_0) A^+_q A_q + (E^B_q - \mu_0) B^+_q B_q \right],
\]

where \( A_q = \alpha_q a_q + \beta_q b_q, \ B_q = \alpha_q b_q - \beta_q a_q, \ \alpha_q^2 + \beta_q^2 = 1, \) and the bare chemical potential \( \mu_0 = g\mu_B (B_c - B). \) The saturation field \( B_c = W/(g\mu_B), \) with \( W \) being the magnon bandwidth, was calculated to be \( B_c = 8.51 \text{ T} \) assuming \( g = 2.20 \) [10, 17].

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, \( n \approx (1 - B/B_c). \) The magnon scattering can be treated in the ladder approximation [18], neglecting interference between \( a \) and \( b \) channels. In this approximation, the problem reduces to solving the Bethe-Salpeter equation in each channel.

This results in the renormalized scattering amplitudes \( \Gamma^{(i)}(\vec{q}_1, \vec{q}_2; \vec{q}_3, \vec{q}_4) \) for \( i = a, b \). Here \( \vec{q}_3, \vec{q}_4 \) and \( \vec{q}_1, \vec{q}_2 \) are magnon momenta before and after scattering, respectively, and \( \vec{q}_3 + \vec{q}_4 = \vec{q}_1 + \vec{q}_2. \) The total energy of scattered magnons was set to zero. This limit is compatible with our main goal to describe the phase transition near \( B_c \) when approaching the phase boundary \( T_c(B) \) from higher temperatures. At \( T \to T_c \), only the magnon states at \( \vec{q} \approx \vec{Q}_{1,2} \) are occupied and the magnon spectrum renormalization near the minima is important.

With given \( \Gamma^{(a)} \) and \( \Gamma^{(b)}, \) the complete set of two-particle scattering amplitudes was then obtained by multiplying \( \Gamma^{(a,b)} \) by products of four \( \alpha_q \) and \( \beta_q \) coefficients. For instance, a scattering process \( (A_{\vec{q}_3}, B_{\vec{q}_4}) \to (A_{\vec{q}_1}, B_{\vec{q}_2}) \) in the channel \( a \) is described by the amplitude \( \alpha_{\vec{q}_3} \beta_{\vec{q}_4} \alpha_{\vec{q}_1} \beta_{\vec{q}_2} \Gamma^{(a)}(\vec{q}_1, \vec{q}_2; \vec{q}_3, \vec{q}_4). \)

The renormalization of low-energy magnons was found by treating the magnon scattering effects in the Hartree-Fock approximation:

\[
H_{\text{int}}^{\text{HF}} = 2\Gamma n \sum_q \left( A^+_q A_q + B^+_q B_q \right) + 2\Gamma' \alpha \beta n \sum_q \left( A^+_q B_q + B^+_q A_q \right),
\]

where \( \alpha_{\vec{q}_1}^2 = \alpha_{\vec{q}_2}^2 = \alpha^2 \) and \( \beta_{\vec{q}_1}^2 = \beta_{\vec{q}_2}^2 = \beta^2. \) Taking into account that \( \alpha^2 \gg \beta^2, \) we keep here only the leading contributions to energy parameters \( \Gamma \) and \( \Gamma': \)

\[
\Gamma \simeq \alpha^4 \Gamma^{(a)}(\vec{Q}_1, \vec{Q}_1; \vec{Q}_1, \vec{Q}_1) = \alpha^4 \Gamma^{(b)}(\vec{Q}_2, \vec{Q}_2; \vec{Q}_2, \vec{Q}_2), \ \Gamma' \simeq 2\Gamma/\alpha^2 \]

and we obtained the estimate \( \Gamma \approx 0.85J. \) According to eq. [5] the chemical potential of magnons is renormalized \( \mu_0 \to \mu_{\text{eff}} = \mu_0 - 2\Gamma n, \) and the low-energy magnons are hybridized due to the second term in eq. [6]. This term shifts the bottom of the magnon band slightly down and leads to a weak mass enhancement of low-energy magnons. Both effects are proportional to \( n^2 \) and we omit them since \( n \ll 1 \) near \( T_c \) for \( (B_c - B) \ll B_c. \)

For a given \( B \ll B_c \) and with decreasing temperature the magnon BEC occurs when the effective chemical potential \( \mu_{\text{eff}} \) vanishes [1]. Then \( T_c(B) \) is determined by

\[
g\mu_B (B_c - B) = 2\Gamma n(T_c).
\]

Here \( n(T) = \sum_q f_B(E_q) \) with \( f_B(E_q) \) being the Bose distribution function taken at \( \mu_{\text{eff}} = 0 \) and \( E_q = E^A_q \) or \( E_q = E^B_q. \) This means that for \( T < T_c \) the magnon condensate develops simultaneously at \( \vec{q} = \vec{Q}_{1,2}. \) It is worth emphasizing that at \( \mu_{\text{eff}} = 0 \) the distribution function \( f_B(E) \) diverges as \( T/E \) for \( E \to 0. \) Therefore, the low energy 3D-magnon spectrum, \( E < E^*, \) mainly contributes and drives the BEC transition. The phase boundary can be calculated using eq. [10]. It gives a very good description of the experimental data near \( B_c \) (see inset to Fig. 4), but deviates strongly at lower fields, i.e., for \( B_c - B > 0.5 \text{ T}. \) 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 \( \phi_{\text{eff}} \approx 1.5 \) close to the predicted value \( \phi_{\text{BEC}} = 3/2 \) characteristic for 3D quadratic dispersion of low-energy magnons [2, 3].

We have presented experimental evidence that in Cs\(_2\)CuCl\(_4\) the field dependence of the critical temperature \( T_c(B) \propto (B_c - B)^{1/\phi} \) close to the critical field \( B_c = 8.51 \text{ T} \) is well described with \( \phi \approx 1.5. \) This is in very good agreement with the exponent expected in the mean-field approximation. Together with the observed opening of a spin gap above \( B_c \) these findings support the notion of a Bose-Einstein condensation of magnons in Cs\(_2\)CuCl\(_4\).

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The nuclear contributions were determined from a plot of $C_{\text{tot}}T^2$ vs $T^3$, assuming that $C_{\text{tot}} = C_{\text{nuc}} + C_{\text{mag}} + C_{\text{ph}}$. The nuclear contributions are due to the hyperfine interactions and can be written as $C_{\text{nuc}} = \alpha/T^2 = (\alpha_Q + \alpha_Z)/T^2$, with the quadrupolar and the Zeeman contribution, respectively. In this way we obtained $\alpha_Q = 33(11) \, \mu\text{JK/mol}$ and $\alpha_Z = 13.8(3) B^2 \, \mu\text{JK/(mol T^2)}$. The latter value is in good agreement with the estimated value $\alpha_Z = 11 \, \mu\text{JK/mol}$, using NMR-data [11].

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