Sound Attenuation Study on the Bose-Einstein Condensation of Magnons in TlCuCl₃

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We investigate experimentally and theoretically sound attenuation in the quantum spin system TlCuCl₃ in magnetic fields at low temperatures. Near the point of Bose-Einstein condensation of magnons a sharp peak in the sound attenuation is observed. The peak demonstrates a hysteresis as a function of the magnetic field pointing to a first-order contribution to the transition. The sound damping has a Drude-like form arising as a result of hard-core magnon-magnon collisions. The strength of the coupling between lattice and magnons is estimated from the experimental data. The puzzling relationship between the transition temperature and the concentration of magnons is explained by their “relativistic” dispersion.

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Quantum spin systems in magnetic fields have recently gained enormous attention because of the richness and universality of phenomena that may be observed [1,2]. The relevance of quantum field theories to quantum spin systems allows the development of theoretical predictions that can be tested experimentally. The observation of plateaus in the high-field magnetization of quantum spin chains predicted in Ref. [3] is an example of this mutual interest. The effect of a magnetic field on a gapped spin system is especially interesting for a small gap, which can be reduced or even closed by experimentally available static fields. On the other hand, sound attenuation experiments were very useful in the investigation of phase transitions with complex order parameters, e.g., superconductivity in the heavy fermion UPt₃ [4] or the movement of flux lines in high- Tc superconductors [5].

The XCuCl₃ compounds, with X = Tl, K, and NH₄, realize especially interesting systems [2]. Magnetization plateaus are observed in NH₄CuCl₃, while TlCuCl₃ and KCuCl₃ show field-induced critical phenomena. The underlying quantum magnetism is based on S = 1/2 spins of Cu²⁺ ions arranged as planar dimers of Cu₂Cl₆. These dimers form infinite double chains parallel to the crystallographic a axis of the monoclinic structure (space group P2₁/c with 4 f.u. per unit cell) [6–8]. The ground state of TlCuCl₃ is a singlet with a spin gap Δ ≈ 7 K and a weakly anisotropic antiferromagnetic intradimer interaction of J ≈ 60 K (Ref. [9]). The magnon dispersion was investigated theoretically in Ref. [10]. The three-dimensional (3D) character of the system leads to a relatively small gap since in this case it is more difficult to bind the magnons. In a magnetic field H the gap for Sₓ = −1 excitations is reduced as Δ − gμBH, where a g factor varies between 2.06 and 2.23 depending on the H direction. Reaching a field of Hₘ = Δ/gμB, the gap is closed.

A possible mechanism of magnetic-field-induced spin transitions introduced in Ref. [11] is the Bose condensation of the soft mode. For the explanation of field-induced ordering in TlCuCl₃ the Bose condensation of diluted magnons with Sₓ = −1 was proposed [12]. A Hartree-Fock description of such a transition leads to an exponent φ = 1.5 relating concentration of magnons nB and the transition temperature Tₜ as nB ∝ Tₜ⁻¹. However, a larger value of φ exp close to 2 has been determined by magnetization and specific heat experiments [12–15].

At the same time, the Bose condensation should be observable in experiments which are directly related to the distribution function of the magnons fB(k), where k is the magnon momentum. Here we present, to our knowledge, the first results of ultrasonic attenuation α(H) measurements on TlCuCl₃ in magnetic field and show that the experimental data suggest a gradual increase of fB(k) in the region of small k with the increase of H being consistent with the Bose-Einsten condensation (BEC) of magnons at a critical field Hc. As we see below, ultrasonic attenuation experiments in this compound allow us to trace the main features of the distribution function, investigate the fluctuation region where the attenuation anomalously increases, make a conclusion on the order of the transition, and determine the mechanism and strength of magnon-lattice coupling.

Ultrasonic attenuation has been measured in single crystals of TlCuCl₃ prepared by the Bridgman method [13] using a pulse-echo method with longitudinal sound waves at a frequency of 5 MHz. The transducers were glued on freshly cleaved or on polished surfaces in the crystallographic (102) plane of the crystals using a liquid polymer (Thiokol, Dow Resin). The H field has been applied parallel to the direction of sound propagation. The value of α(H) is determined from the experimental data as α(H) = 10log₁₀(I₀/Iₙ+₁), where I₀ and Iₙ+₁ are the intensities of Nth and Nth + 1 pulses arriving at the detector with the time interval 2tp, where tp is the pulse traveling time through the sample.
The experimental results shown in Fig. 1 can be summarized as the following observations:

(i) a weak $H$ dependence up to $H = 2.5$ T,

(ii) a sharp and asymmetric peak close to the critical field $H_c$, determined by thermodynamic experiments [12]. The sharp peak associated with the fluctuation region of the transition shows a pronounced hysteresis with increasing/decreasing field, which indicates a first-order contribution to the transition, and

(iii) a decrease of $\alpha(H)$ for $H > 9.7$ T.

If the comparably slow underlying increase is approximated by a Gaussian, the sharp peak near $H_c$ can be separated from the background. Inset (b) shows the resulting transition-induced attenuation at three different temperatures. The peaks have a gradual low-field onset of attenuation $H_c^{\text{ons}}$, a sharp maximum $H_c^{\text{max}}$, and a high-field cutoff $H_c^{\text{co}}$. The two critical fields $H_c^{\text{ons}}$ and $H_c^{\text{co}}$ show a pronounced hysteresis. The respective attenuation at the critical fields systematically changes with $T$. For $H_c^{\text{ons}}$, which marks a crossover into a regime with strong fluctuations, the hysteresis is negligible. In a recent NMR investigation a hysteresis close to the phase boundary has also been observed and attributed to spin-phonon coupling [16]. Inset (c) shows the resulting phase diagram compared to magnetization measurements [12] (thick dashed line).

The energy of a propagating sound pulse decays with time as $w(t) \sim \exp[-(1 + \gamma_m(H))t]$, and, therefore $\alpha(H) = 20\log_{10}e \cdot (1 + \gamma_m(H))t$. Here $\gamma_m(H)$ is the contribution of magnons coupled to the lattice, while an unknown background $\Gamma$ arises due to all other effects such as scattering by impurities, interfaces, etc. In contrast with $\Gamma$, the $H$ dependence of the damping $\gamma_m(H) - \gamma_m(0)$ which corresponds to the field-induced changes in $f_B(k)$ and sound attenuation mechanism can be determined accurately from Fig. 1. By comparison with theoretical calculations also $\gamma_m(0)$ will be obtained.

Cottam [17] investigated spin-phonon coupling in an antiferromagnet in the case of a low density of magnons where magnon-magnon interactions can be neglected. Below we investigated another case where the damping is related to collisions between magnons. The model for sound attenuation is the following: In the field of the longitudinal sound wave with wave vector $q$ and frequency $\omega$, $u(r) = u_0 \exp[i(qr - \omega t)]$, leads to an effective time-dependent force $F = -\nabla \delta E(k)$ with $F = C_k u_0 q^2$ driving changes in the velocity of magnons. Since one cannot create two $S_z = -1$ states on a Cu-Cu bond, the magnons collide like hard-core bosons. Inelastic collisions between the magnons driven by the displacements of the lattice in the wave lead to a dissipation of the pulse energy. The dissipation rate per one magnon is equal to the average of the product $F \nu$ per period, where $\nu$ is the magnon velocity. The sound dissipation rate in this case is the dissipation rate of an external harmonic field in a medium, generally described by a Drude-like mechanism of the form

$$\gamma_m(H) = \frac{A u_0^4 q^3 (n\sigma)^{-1}}{(2\pi)^4} \frac{1 + \omega^2/(\tau)^2}{1 + \omega^2/(\tau)^2} \int d^3 k f_B(k) \frac{C_k^2}{m_k} \int d^3 k' f_B(k') \times \frac{1}{m_k \nu_{kk'}},$$

where $m_k$ is the magnon effective mass and $\nu_{kk'}$ is the relative velocity of the particles with momenta $k$ and $k'$ that determines their collision rate. The constant $\sigma$ is of the order of 1 and depends on details of magnon-magnon collisions, which are beyond our consideration. The mean time between the collisions $\langle \tau \rangle \sim (\nu^{-1})$ with the magnon free path $\ell$ being proportional to $1/n\sigma$, where $\sigma$ is the magnon-magnon scattering cross section, and $n$ is the magnon concentration, is, therefore sensitive to the distribution function. The mean value $\langle \nu^{-1} \rangle$ and the damping increase simultaneously. Since $\omega(\tau)$ is very small ($\sim 0.01$) at the experimental $\omega \sim 3 \times 10^7$ s$^{-1}$, it is neglected in the denominator of Eq. (1) for the rest of the discussion.

To include the specific spin excitation spectrum, we start with the “relativistic” form of the magnon dispersion $\epsilon(k) = \sqrt{\Delta^2 + J_2 k^2}/4$ at small $k$ which is important if $T_B$ and $\Delta$ are of the same order of magnitude as in TICuCl$_3$. From here on the lattice constant $l = 1$ if otherwise is not stated. Since thermal $k \ll 1$ at $T_B \ll J$ this form of energy is sufficient for an understanding of the Bose condensation in the experimental temperature range. The velocity of a magnon and its inverse mass are (we put $\hbar = 1$)

$$v(k) = \frac{d\epsilon(k)}{dk} = \frac{J^2}{4\epsilon(k)} k, \quad \frac{1}{m_k} = \frac{J^2}{4} \left[ \frac{1}{\epsilon(k)} - \frac{J^2}{4} \frac{\epsilon^3(k)}{\epsilon(k)} \right].$$

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**FIG. 1.** (a) Ultrasonic attenuation $\alpha(H)$ with increasing (full circles) and decreasing magnetic field (open circles) and a fitting of the broad maximum by a Gaussian (dashed line). Inset (b): transition regime after subtracting the Gaussian and a constant. Inset (c): phase diagram with $gH_c/2$ (thick line) from thermodynamic experiments [12]. The $g$ factor of $S_z = -1$ excitations $g = 2.16$. 

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respectively. The mass at small $k$, $m = 4\Delta/J^2$ determines $T_B$ at low concentration of magnons where $T_B \ll \Delta$. In the mean-field approximation which is used below, the distribution function is $f_B(\varepsilon) = 1/[\exp((\varepsilon - \mu_{\text{eff}})/T) - 1]$, where $\varepsilon(k) = \varepsilon(k) - \Delta$, and $\mu_{\text{eff}} = g\mu_B(H - H_\text{c}) - 2V_{\text{mm}}n$. $n$ is the dimensionless concentration of the magnons determined by the magnetization $M$ as $n = M/H$, and $V_{\text{mm}}$ is the magnon-magnon interaction parameter. The transition temperature $T_B$ obtained with the condition $\mu_{\text{eff}} = 0$, that is

$$4\pi\int \frac{k^2}{\varepsilon(k)/T_B - 1} \frac{dk}{(2\pi)^3} = n_B.$$  \hspace{1cm} (3)

is, therefore, $V_{\text{mm}}$ independent. Figure 2(a) presents dimensionless $n_B(T_B)$, and Fig. 2(b) shows the ratio $[\pi_B(T_B)/\pi_B(1 \text{ K})]/T_B^2$ obtained from Eq. (3) for different $\Delta$ and $J = 7\Delta$. As one can see, at the chosen $\Delta = 6.5 \text{ K}$, the ratio is nearly a constant in the experimental range of temperatures, giving an explanation for the observation of $\phi_{\exp} = 2$ [12–15]. Since near the condensation point the momenta of the magnons $k^2 \sim mT_B$, leading to a temperature dependence of the mean value $\langle \varepsilon(k) - \Delta \rangle - T$, the “exponent” depends on $T_B/\Delta$ only. We note that $\phi_{\exp}$ is indeed a fitting parameter, which can depend on the measured quantity. We mention that the parameter which determines the applicability of the Drude approach $k \ell$ and can be estimated as $\propto n^{-2/3}$, where $k$ is the characteristic magnon momentum, is always larger than 10 thus justifying the validity of Eq. (1) except the fluctuation region.

Since near the transition the Bose function behaves as

$$f_B(\varepsilon) = \frac{T_B}{\varepsilon - |\mu_{\text{eff}}|},$$  \hspace{1cm} (4)

due to the singularity at $\mu_{\text{eff}} \to 0$, the average $\langle n^{-1} \rangle$ diverges as $\ln(T_B/|\mu_{\text{eff}}|)$ leading, as is shown below, to an increase in the damping rate.

The sound-induced change in the magnon energy consists of two terms: $\delta\varepsilon(k) = \delta\varepsilon_{\Delta}(k) + \delta\varepsilon_{\text{J}}(k)$ arising due to a modulation in the gap $\Delta = \Delta + C_{\text{J}}\nabla u$ and the exchange $J = J + C_{\text{J}}\nabla u$, respectively. The resulting deformation potential is

$$C_{\text{J}} = \frac{\Delta}{\varepsilon(k)}C_{\Delta} + \frac{J}{\varepsilon(k)} \frac{k^2}{4} C_{\text{J}}.$$  \hspace{1cm} (5)

Since the $J$-originated term in $C_{\text{J}}$ vanishes at small $k$, the main contribution to the increase of damping near the transition comes from $C_{\Delta}$. The calculated behavior of the damping at $T = 3.5 \text{ K}$ for $H < H_\text{c}$ is shown in Fig. 3 for two $V_{\text{mm}}$ and two models of phonon-magnon coupling. As a result of the increase of $f_B(k)$ at small $k$ near the transition point, the damping due to the $C_{\Delta}$ term increases and shows a $\gamma_{\text{m}}(H)$ behavior which is in full agreement with the experimental data in Fig. 1. At the same time, due to a vanishing coupling for $k \to 0$ in the second contribution in Eq. (5), the $C_{\text{J}}$-dependent term leads to a slight decrease in the damping, in contrast to Fig. 1. Moreover, near the transition point, where $k^2 \sim T_B^2/\Delta^2$, the ratio of the $C_{\Delta}$ and $C_{\text{J}}$-originated terms in Eq. (5) is of the order of $C_{\text{J}}T_B^2/C_{\Delta}$, thus containing a small factor of $T_B^2/\Delta$ that decreases the role of the $C_{\text{J}}$-originated term.

Our approach can qualitatively explain the decrease in the damping rate when the field exceeds some value corresponding to the maximum at $H = 9.7 \text{ T}$ in Fig. 1. The above consideration is based on the low-energy

![FIG. 2. (a) Dimensionless concentration of magnons $n_B(T_B)$ corresponding to the BEC temperature $T_B$. Squares present experimental data [12]. (b) $[\pi_B(T_B)/\pi_B(1 \text{ K})]/T_B^2$. With increasing $\Delta$ the curves are closer to $T_B^{1/2}$ (the dashed line), corresponding to $\phi = 3/2$.](image)

![FIG. 3. Calculated $\mu_{\text{eff}}$ (a) and damping (b),(c) for different spin-lattice couplings as a function of field $H$ at $T = 3.5 \text{ K}$. For $H < 2.5 \text{ T}$ the behavior of damping is almost flat, in agreement with the experiment. $\Delta = 6.5 \text{ K}, H_\text{c} = 4.5 \text{ T}$. The results depend on $V_{\text{mm}}$ which determines the $H_\text{c}$.](image)
The dissipation rate estimated from Eq. (1) is $V_{hMM} \sim 3m$, and, therefore, $\gamma_m(0) \sim 0.2 \times 10^4 \text{ s}^{-1}$. The density $\rho = 5 \text{ g/cm}^3$, $\omega = 3 \times 10^7 \text{ s}^{-1}$, and $l = \Omega^{1/3} = 5 \text{ A}$, where $\Omega$ is the unit cell volume per chemical formula yield $C_A$ of the order of 200 K.

We presented ultrasonic attenuation experiments as well as a theoretical analysis on the field-induced changes in the spin dimer system TiCuCl$_4$. These experiments are in agreement with the magnon Bose-Einstein condensation and summarized as a sharp peak in the attenuation $\alpha(H)$ in the close vicinity of the transition and a second broader maximum at larger fields, deep in the long-range ordered phase. The peak in $\alpha(H)$ shows a hysteresis crossing the phase boundary with increasing/decreasing field. This effect is attributed to a first-order contribution to the phase transition. The exponent $\phi = 2$ in the relation $n_B \sim T^\phi_f$ and the $H$ dependence of sound attenuation originate from the relativistic dispersion of magnons. The spin-lattice coupling is due to a phonon-induced modulation of the spin gap with the coupling constant $C_A \sim 200 \text{ K}$. The recent observation [19,20] of triplet crystallization in the 2D compound SrCu$_2$(BO$_3$)$_2$ in large magnetic fields with similar spin-phonon coupling demonstrates that these phenomena are of a general nature.

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