Sound Attenuation Study on the Bose-Einstein Condensation of Magnons in $TlCuCl_3$

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We investigate experimentally and theoretically sound attenuation in the quantum spin system $TlCuCl_3$ 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 UPt_3 [4] or the movement of flux lines in high-T_c superconductors [5].

The $X$$CuCl_3$ compounds, with $X = $ Tl, K, and NH_4, realize especially interesting systems [2]. Magnetization plateaus are observed in NH_4CuCl_3, while $TlCuCl_3$ and $KCuCl_3$ show field-induced critical phenomena. The underlying quantum magnetism is based on $S = 1/2$ spins of Cu^{2+} ions arranged as planar dimers of Cu_2Cl_6. These dimers form infinite double chains parallel to the crystallographic a axis of the monoclinic structure (space group $P2_1/c$ with 4 f.u. per unit cell) [6–8]. The ground state of $TlCuCl_3$ is a singlet with a spin gap $\Delta \approx 7$ K and a weakly anisotropic antiferromagnetic intradimer interaction of $J \sim 60$ 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 $S_z = -1$ excitations is reduced as $\Delta - g\mu_B H$, where a g factor varies between 2.06 and 2.23 depending on the $\mathbf{H}$ direction. Reaching a field of $H_g = \Delta/g\mu_B$, 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 magnetic ordering in $TlCuCl_3$ the Bose condensation of diluted magnons with $S_z = -1$ was proposed [12]. A Hartree-Fock description of such a transition leads to an exponent $\phi = 1.5$ relating concentration of magnons n_B and the transition temperature T_B as $n_B \sim T_B^{\phi}$. However, a larger value of $\phi_{\rm exp}$ 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 $f_B(k)$, where k is the magnon momentum. Here we present, to our knowledge, the first results of ultrasonic attenuation $\alpha(H)$ measurements on $TlCuCl_3$ in magnetic field and show that the experimental data suggest a gradual increase of $f_B(k)$ in the region of small k with the increase of H being consistent with the Bose-Einstein condensation (BEC) of magnons at a critical field H_c. 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 $TlCuCl_3$ 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 $(10\bar{2})$ plane of the crystals using a liquid polymer (Thiokol, Dow Resin). The $\mathbf{H}$ field has been applied parallel to the direction of sound propagation. The value of $\alpha(H)$ is determined from the experimental data as $\alpha(H) = 10\log_{10}(I_N/I_{N+1})$, where I_N and I_{N+1} are the intensities of Nth and Nth + 1 pulses arriving at the detector with the time interval $2t_p$, where t_p 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 $H \approx 2.5$ 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 $H_c^{\rm ons}$, a sharp maximum $H_c^{\rm max}$, and a high-field cutoff $H_c^{\rm co}$. The two critical fields $H_c^{\rm max}$ and $H_c^{\rm co}$ show a pronounced hysteresis. The respective attenuation at the critical fields systematically changes with T. For $H_c^{\rm ons}$, 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 $w(t) \sim \exp\{-[\Gamma + \gamma_m(H)]t\}$, and, therefore $\alpha(H) = 20\log_{10}e \cdot [\Gamma + \gamma_m(H)]t_p$. Here $\gamma_m(H)$ is the contribution of magnons coupled to the lattice, while an unknown background Γ arises due to all other effects such as scattering by impurities, interfaces, etc. In contrast with Γ, 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.

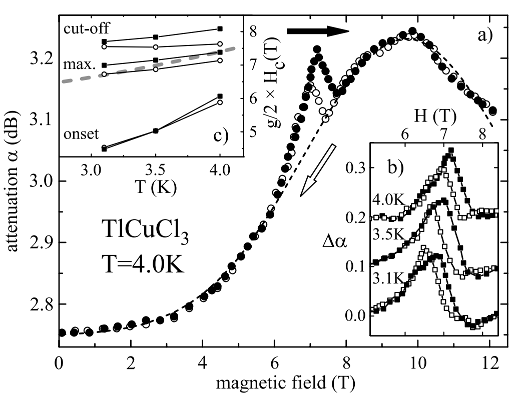

FIG. 1. (a) Ultrasonic attenuation $\alpha(H)$ with increasing (full circles) and decreasing magnetic field (open circles) and a fitting of the broad maximum by a Gaussian (dashed line). Inset (b): transition regime after subtracting the Gaussian and a constant. Inset (c): phase diagram with $gH_c/2$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

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 displacements of ions $\mathbf{u}(\mathbf{r})$, the energy of the magnon changes due to magnon-lattice coupling as $\delta\varepsilon(\mathbf{k}) = C_k \nabla \mathbf{u}(\mathbf{r})$, where C_k is the deformation potential. The spatial dependence of the displacement in a plane wave with wave vector q and frequency ω, $\mathbf{u}(\mathbf{r}) = \mathbf{u}_0 \exp[i(\mathbf{qr} - \omega t)]$, leads to an effective time-dependent force $\mathbf{F} = -\nabla\delta\varepsilon(\mathbf{k})$ 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 $\mathbf{Fv}$ per period, where $\mathbf{v}$ 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

$$\gamma_m(H) = \frac{Au_0^2 q^4}{(2\pi)^6} \frac{(n\sigma)^{-1}}{1 + \omega^2 \langle\tau\rangle^2} \int d^3k f_B(k) \frac{C_k^2}{m_k} \int d^3k' f_B(k') \times \frac{1}{n v_{\mathbf{k},\mathbf{k}'}}, \tag{1}$$

where m_k is the magnon effective mass and $v_{\mathbf{k},\mathbf{k}'}$ is the relative velocity of the particles with momenta $\mathbf{k}$ and $\mathbf{k}'$ that determines their collision rate. The constant A 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 $\langle\tau\rangle \sim \langle \ell v^{-1}\rangle$ with the magnon free path ℓ being proportional to $1/n\sigma$, where σ is the magnon-magnon scattering cross section, and n is the magnon concentration, is, therefore sensitive to the distribution function. The mean value $\langle v^{-1}\rangle$ and the damping increase simultaneously. Since $\omega\langle\tau\rangle$ is very small (~ 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 $\varepsilon(k) = \sqrt{\Delta^2 + J^2k^2/4}$ at small k which is important if T_B and Δ are of the same order of magnitude as in $TlCuCl_3$. From here on the lattice constant $l \equiv 1$ 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 $\hbar \equiv 1$)

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

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 $f_B(\varepsilon) = 1/\{\exp[(\bar{\varepsilon} - \mu_{\rm eff})/T] - 1\}$, where $\bar{\varepsilon}(k) = \varepsilon(k) - \Delta$, and $\mu_{\rm eff} = g\mu_B(H - H_g) - 2V_{\rm mm}\bar{n}$. $\bar{n}$ is the dimensionless concentration of the magnons determined by the magnetization M as $\bar{n} = M/H$, and $V_{\rm mm}$ is the magnon-magnon interaction parameter. The transition temperature T_B obtained with the condition $\mu_{\rm eff} = 0$, that is

$$4\pi \int \frac{k^2}{e^{\bar{\varepsilon}(k)/T_B} - 1}\frac{dk}{(2\pi)^3} = n_B, \tag{3}$$

is, therefore, $V_{\rm mm}$ independent. Figure 2(a) presents dimensionless $\bar{n}_B(T_B)$, and Fig. 2(b) shows the ratio $[\bar{n}_B(T_B)/\bar{n}_B(1\ {\rm K})]/T_B^2$ obtained from Eq. (3) for different Δ and $J = 7\Delta$. As one can see, at the chosen $\Delta = 6.5$ K, the ratio is nearly a constant in the experimental range of temperatures, giving an explanation for the observation of $\phi_{\rm exp} \approx 2$ [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 $\langle \varepsilon(k) - \Delta \rangle \sim T$, the "exponent" depends on T_B/Δ only. We note that $\phi_{\rm exp}$ 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 $\sim\bar{n}^{-2/3}$, 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

$$f_B(\varepsilon) = \frac{T_B}{\bar{\varepsilon} + |\mu_{\rm eff}|}, \tag{4}$$

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

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

$$C_k = \frac{\Delta}{\varepsilon(k)}C_\Delta + \frac{J}{\varepsilon(k)}\frac{k^2}{4}C_J. \tag{5}$$

Since the J-originated term in C_k vanishes at small k, the main contribution to the increase of damping near the transition comes from C_Δ. The calculated behavior of the damping at $T = 3.5$ K for $H < H_c$ is shown in Fig. 3 for two $V_{\rm mm}$ 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_Δ term increases and shows a $\gamma_m(H)$ 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_J-dependent term leads to a slight decrease in the damping, in contrast to Fig. 1. Moreover, near the transition point, where $k^2 \sim T_B\Delta/J^2$, the ratio of the C_J- and C_Δ-originated terms in Eq. (5) is of the order of $C_J T_B/JC_\Delta$, thus containing a small factor of T_B/J that decreases the role of the C_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 $H = 9.7$ T in Fig. 1. The above consideration is based on the low-energy

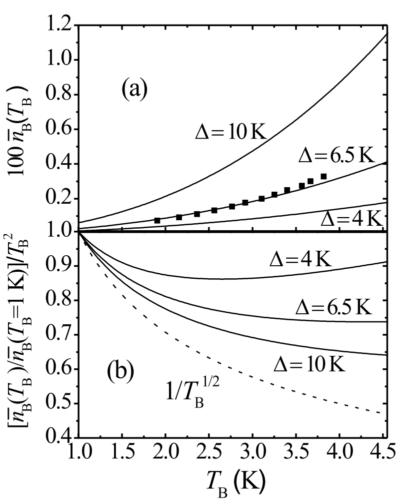

FIG. 2. (a) Dimensionless concentration of magnons $\bar{n}_B(T_B)$ corresponding to the BEC temperature T_B. Squares present experimental data [12]. (b) $[\bar{n}_B(T_B)/\bar{n}_B(1\ {\rm K})]T_B^{-2}$. With increasing Δ the curves are closer to $T_B^{-1/2}$ (the dashed line), corresponding to $\phi = 3/2$.

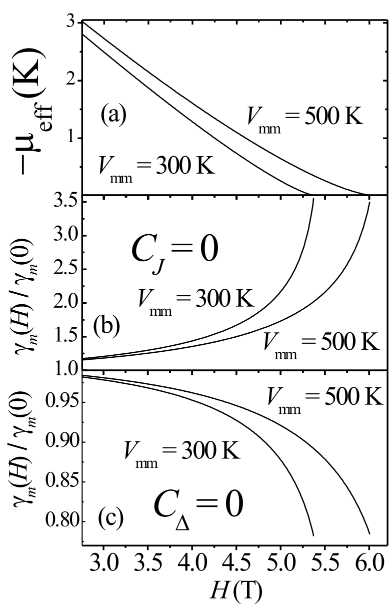

FIG. 3. Calculated $\mu_{\rm eff}$ (a) and damping (b),(c) for different spin-lattice couplings as a function of field H at $T = 3.5$ K. For $H < 2.5$ T the behavior of damping is almost flat, in agreement with the experiment. $\Delta = 6.5$ K, $H_g = 4.5$ T. The results depend on $V_{\rm mm}$ which determines the H_c.

singularity of the $f_B(\varepsilon)$ increasing as $1/\overline{\varepsilon}$ at $H = H_c$. At $H > H_c$ the number of quasiparticles $\overline{n}$ rapidly increases with H [12]. The spectrum of excitations in this case is $E(k) = v_c k$, with $v_c \sim \overline{n}^{1/2}$ being the condensate sound velocity. With the increase of H, and, correspondingly $\overline{n}$, the excitation energy increases, and, therefore, the role of thermal excitations decreases. Thus, the increase in H drives the condensate effectively towards the zero-temperature behavior. The system demonstrates a zero-T behavior when $\overline{n}$ becomes much larger than n_B corresponding to the BEC at given T. At zero T the distribution of the out-of-condensate particles which contribute to the damping at small momenta is $N_E(E \to 0) = mv_c^2/2E(k)$ [18]. Since $E(k)$ is linear in k, the singularity in Eq. (4) at $\mu_{\mathrm{eff}} = 0$ becomes weaker, and the damping decreases. To estimate the field at which the behavior of the condensate is close to the $T = 0$ limit we note that at $T > 0$ thermal excitations and interaction V_{mm} produce out-of-condensate particles. The concentration of the magnons at which the system demonstrates a crossover to the $T = 0$ behavior is determined by the condition that the concentration of the particles out of the condensate arising due the interaction ($\sim n\sqrt{na^3}$) is larger than the concentration of thermally excited particles proportional to $T^{3/2}$, where $a = V_{\mathrm{mm}} l^3 m/4\pi$ is the magnon-magnon scattering length. In $\mathrm{TlCuCl_3}$ a and l are close to each other since the hard-core boson has a spatial size of the Cu-Cu distance. As a result, the critical concentration which determines the crossover to zero-temperature behavior and, in turn, the broad maximum position is $\overline{n}_{\mathrm{cr}} \sim \overline{n}_B(l/a\overline{n}_B^{1/3}) \sim 10\overline{n}_B$. On the other hand, from the data on $n(H)$ dependence (Ref. [12]) one expects that at $T = 3.5$ K, $\overline{n}(H = 10\ \mathrm{T})$ is an order of magnitude larger than $\overline{n}_B(H_c)$, in a good qualitative agreement with our estimate.

Following Eqs. (1) and (5) one can estimate C_Δ. The energy density in the sound wave with $\omega = cq$ (c is the sound velocity) is $w \sim \kappa u_0^2 \omega^2/c^2$, where κ is the elastic constant. The dissipation rate estimated from Eq. (1) is

$$\frac{dw}{dt} \sim \frac{F^2}{m}\frac{1}{\sigma\langle v\rangle} \sim C_\Delta^2 \frac{\omega^4}{c^4} u_0^2 \frac{1}{m\sigma\langle v\rangle}. \qquad (6)$$

Therefore, the relaxation rate $(dw/dt)w^{-1} \sim \omega^2$. Let us consider as an example $\gamma_m(0)$ that with the estimates given above can be written as

$$\gamma_m(0) \sim \frac{dw}{dt}\frac{1}{w} \sim \frac{C_\Delta^2}{\rho c^2}\frac{\omega^2}{c^2}\frac{1}{l}\frac{J}{\sqrt{\Delta T}}, \qquad (7)$$

where we took into account that far from the transition point $\langle 1/v\rangle \sim 1/\langle v\rangle$ with $\langle v\rangle \sim \sqrt{T/m}$. From the theoretical $\gamma_m(H)$ dependence (Fig. 3), we conclude that $\gamma_m(H_g)/\gamma_m(0) \sim 2$. From the experimental data in Fig. 1 we obtain $\gamma_m(H_g) - \gamma_m(0) \sim 0.1$ dB, and, therefore, $\gamma_m(0)$ corresponds to a damping of the order of 0.1 dB. For the thickness of the sample of 2 mm and the sound velocity $c = 3 \times 10^5$ cm/s, the pulse traveling time t_p is 6.6×10^{-7} s, and, therefore, $\gamma_m(0) \sim 0.2 \times 10^4\ \mathrm{s^{-1}}$. The density $\rho = 5\ \mathrm{g/cm^3}$, $\omega \sim 3 \times 10^7\ \mathrm{s^{-1}}$, and $l = \Omega^{1/3} \approx 5$ Å, where Ω is the unit cell volume per chemical formula yield C_Δ 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 $\mathrm{TlCuCl_3}$. 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 $\phi \approx 2$ in the relation $n_B \sim T_B^\phi$ 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 $C_\Delta \sim 200$ K. The recent observation [19,20] of triplet crystallization in the 2D compound $\mathrm{SrCu_2(BO_3)_2}$ in large magnetic fields with similar spin-phonon coupling demonstrates that these phenomena are of a general nature.

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