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Neutron Scattering Study of the Field-Induced Néel Ordering and Bose-Einstein Condensation of Magnons in $TlCuCl_3$

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Neutron elastic scattering experiments have been carried out on the spin gap system $TlCuCl_3$ in magnetic fields parallel to the b-axis. The magnetic Bragg peaks indicative of the field-induced Néel ordering were observed for magnetic field $H > H_g \approx 5.6$ T in the $a^* - c^*$ plane. The spin structure in the ordered phase was determined and the transverse spin ordering was confirmed. The Bragg peak intensities were measured as a function of temperature and magnetic field. The present results are discussed in connection with a theory of the Bose-Einstein condensation of magnons [Nikuni *et al.*: Phys. Rev. Lett. **84** (2000) 5868.].

KEYWORDS: $TlCuCl_3$, spin gap, neutron elastic scattering, field-induced transverse Néel ordering, Bose-Einstein condensation, magnons

§1. Introduction

In this note we will show the evidence of the field-induced transverse magnetic ordering in the spin gap system $TlCuCl_3$. This compound has a monoclinic structure (space group $P2_1/c$).[1] The crystal structure consists of planar dimer of Cu_2Cl_6. The dimers are stacked to form infinite double chains along the crystallographic a-axis. These chains are located at the corners and centre of the unit cell in the $b - c$ plane, and are separated by Tl^+ ions. The magnetic interaction in $TlCuCl_3$ is described by the $S = 1/2$ Heisenberg model. The magnetic ground state is the spin singlet with an excitation gap $\Delta/k_B \approx 7.5$ K.[2,3] The lowest excitation which corresponds to the gap occurs at $Q = (0, 0, 1)$ and its equivalent reciprocal points,[4] as observed in $KCuCl_3$.[5,6] The origin of the gap is the strong antiferromagnetic interaction on the planar dimer Cu_2Cl_6 in the double chain. The neighboring dimers couple magnetically along the chain and in the $(1, 0, -2)$ plane.

By the magnetization measurements, it was found that $TlCuCl_3$ undergoes 3D magnetic ordering in magnetic fields higher than the critical field $H_g = \Delta/g\mu_B \approx 5.6$ T.[3] The magnetization displays a cusplike minimum at the Néel temperature T_N. The phase boundary is independent of the field direction when normalized by the g-factor, and can be represented by the power law. These features cannot be explained by the mean-field theory.[7]

Nikuni *et al.*[8] argued that field-induced magnetic ordering in $TlCuCl_3$ can be represented as a Bose-Einstein condensation (BEC) of excited triplets (magnons). In the magnon BEC theory, the magnetization M is expressed by the number of magnons N as $M = g\mu_B N$. The observed temperature dependence of the magnetization and the field dependence of the Néel temperature,[3] were qualitatively well described by the magnon BEC theory.[8]

When the magnons undergo BEC at ordering vector Q_0 for $H > H_g$, long-range order for the transverse spin components characterized by the same wave vector Q_0 can occur. The transverse magnetization $m_\perp$ per site is expressed by the condensate density n_c as

$$m_\perp = g\mu_B\sqrt{n_c/2}\ , \qquad (1)$$

when the magnons are dilute. Thus the condensate density n_c is proportional to the magnetic Bragg peak intensity.

In order to confirm the transverse spin ordering and to investigate the temperature and field dependence of the transverse magnetization $m_\perp$ corresponding to the condensate density n_c, we carried out neutron elastic scattering experiments on $TlCuCl_3$.

§2. Experiments

Neutron scattering experiments were performed at the E1 spectrometer of the BER II Research Reactor of the Hahn-Meitner Institute with the vertical field cryomagnet VM1. The incident neutron energy was fixed at $E_i = 13.9$ meV, and the horizontal collimation sequence was chosen as 40'-80'-40'. A single crystal of ~ 0.4 cm^3 was used. The sample was mounted in the cryostat with its (0, 1, 0) cleavage plane parallel to the scattering plane, so that the reflections in the $a^* - c^*$ plane were investigated. The sample was cooled to 0.05 K using a dilution refrigerator. An external magnetic field of up to 12 T was applied along the b-axis.

§3. Results and Discussions

Magnetic Bragg reflections were observed for $H > H_g \approx 5.6$ T, and can be indexed by $Q = (h, 0, l)$ with odd l. These reciprocal lattice points are equivalent to those for the lowest magnetic excitation at zero field.[4] This result indicates that the magnetic unit cell is the same as the chemical one. Ferromagnetic Bragg reflections due to the induced moment could not be detected in the present measurements. The integrated intensities of nine magnetic reflections were measured at 2 K under an external field of 12 T. The results are summarized in Table I together with the calculated intensities.

The spin structure is determined as shown in Fig. 1 at 2K and 12 T. Spins lie in the $a-c$ plane which is perpendicular to the applied field. Spins on the same dimers represented by thick lines in Fig. 1 are antiparallel. Spins are arranged in parallel along a leg in the double chain, and make an angle of 39° with the a-axis. Thus the field-induced transverse magnetic ordering as predicted by the theory[7,8] was confirmed by the present experiment.

We evaluate the magnitude of the staggered perpendicular magnetization as $m_\perp = g\mu_B\langle S_\perp\rangle = 0.26(2)$ μ_B at 2K and 12 T by comparing magnetic peak intensities with those of nuclear reflections, which were corrected for the extinction effect. For $T \to 0$, the condensate density n_c approximates the density of the magnon n, which is given by $n = 2m/g\mu_B$ with the magnetization m per site. The value of n at 12 T and 1.8 K is obtained as $n \approx 0.028$ with $m \approx 0.03$ μ_B and $g = 2.06$.[2,3] Substituting $n_c \approx n \approx 0.028$ into eq. (1), $m_\perp \approx 0.24$ μ_B. This value agrees with $m_\perp = 0.26(2)$ μ_B obtained by the present measurement.

Table I. Observed and calculated magnetic Bragg peak intensities at $T = 2$ K and $H = 12$ T for $H \| b$. The intensities are normalized to $(0, 0, 1)_M$ reflection.

(h,k,l)	I_{obs}	I_{cal}
$(0, 0, 1)_M$	1	1
$(0, 0, 3)_M$	0	0.014
$(0, 0, 5)_M$	0.063	0.107
$(1, 0, 1)_M$	0.031	0.009
$(1, 0, -1)_M$	0.074	0.127
$(1, 0, 3)_M$	0.056	0.024
$(1, 0, -3)_M$	0.398	0.380
$(2, 0, 1)_M$	0.012	0.013
$(2, 0, -1)_M$	0.128	0.102

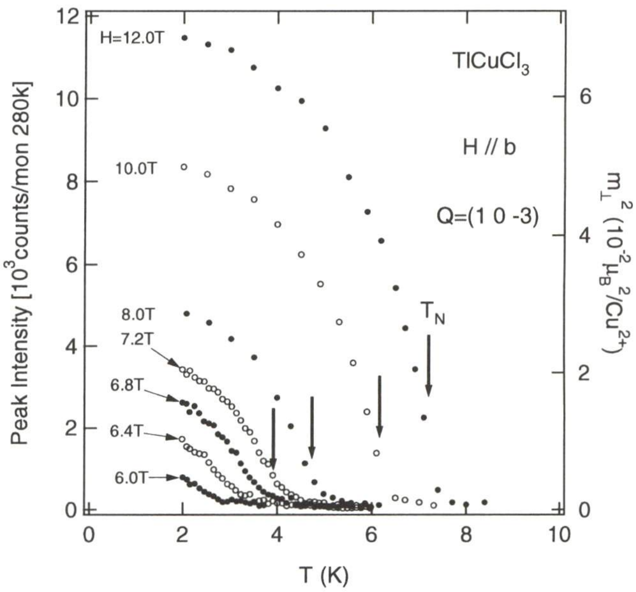

Fig. 2. Temperature dependences of the Bragg peak intensity for $(1, 0, -3)$ reflection and the square of the transverse magnetization per site $m_\perp^2$ at various magnetic fields.

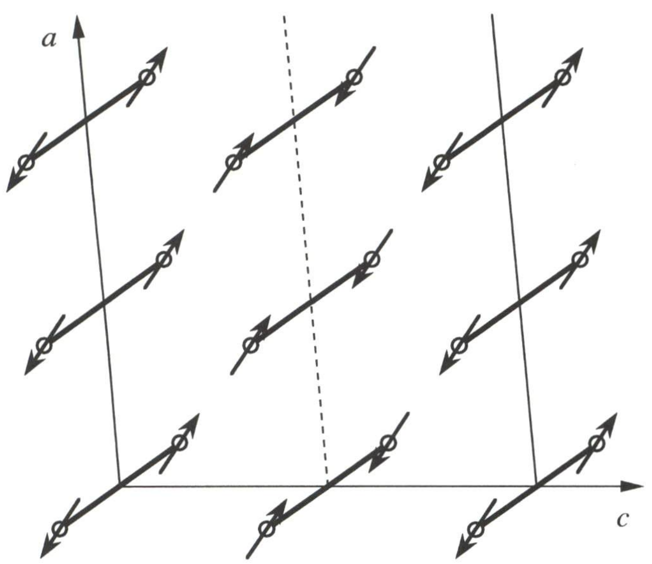

Fig. 1. Spin structure in the ordered phase of $TlCuCl_3$. The external field is applied along the b-axis. The double chain located at the corner and the center of the chemical unit cell in the $b-c$ plane are represented by solid and dashed lines, respectively.

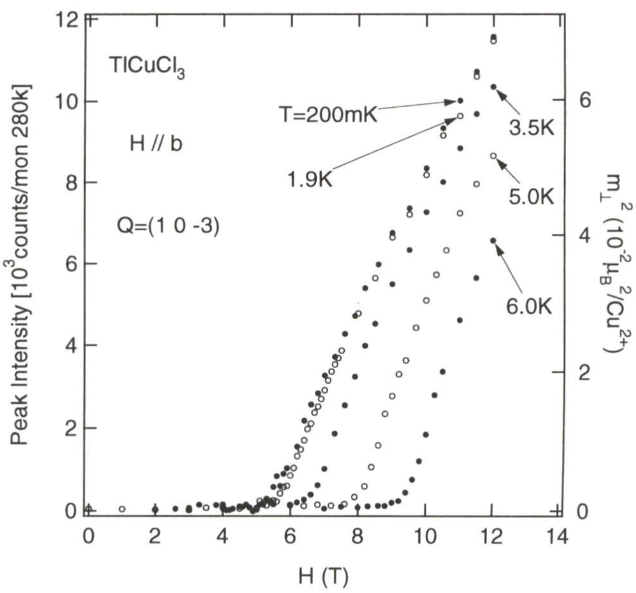

Fig. 3. Magnetic field dependences of the Bragg peak intensity for $(1, 0, -3)$ reflection and the square of the transverse magnetization per site $m_\perp^2$ at various magnetic fields.

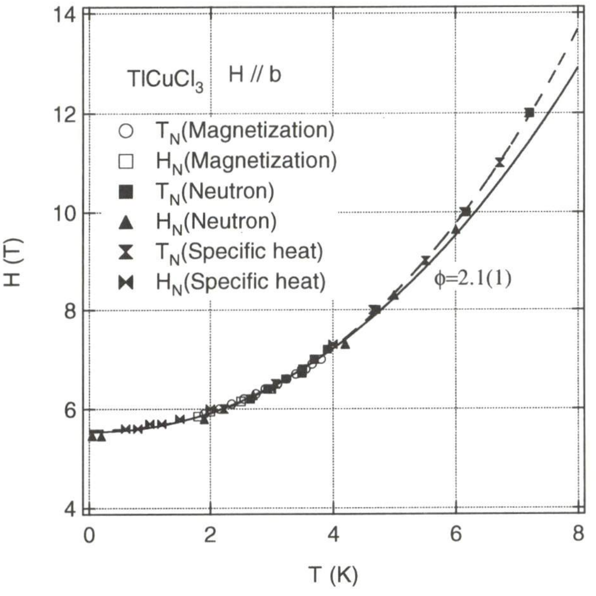

Fig. 4. The phase boundary in $TlCuCl_3$ for $H\|b$ determined from the results of the temperature variation (closed rectangles) and field variation (closed circles) of the Bragg peak intensity. The phase transition data obtained from the magnetic measurements and the specific heat measurements are also plotted. The dashed line is guide for the eyes. The solid line denotes the fit by eq. (2) with $\phi = 2.1(1)$.

Figure 2 shows the temperature dependence of the magnetic peak intensity for $(1, 0, -3)$ measured at various magnetic fields. On the right axis, we show $m_\perp^2$. The Néel ordering can be detected at the temperatures indicated by the arrows. With increasing external field, the Néel temperature $T_N(H)$ and the intensity of the magnetic Bragg peak increase. The anomaly around $T_N(H)$ becomes smeared as the magnetic field approaches H_g. Since the sharp anomaly is observed at the transition field $H_N(T)$ in the field dependence of the Bragg intensity (see Fig. 3), and the sharp phase transition was detected through the specific heat measurement,[9] the smearing of the peak intensity around $T_N(H)$ is attributed not to the intrinsic smearing of the ordering temperature due to additional interaction, but to the diffuse scattering due to the spin fluctuation. The amount of diffuse scattering around $T_N(H)$ seems to be independent of peak intensity at $T = 0$.

Figure 3 shows the field dependences of the Bragg intensity for $(1, 0, -3)$ reflection and $m_\perp^2$ measured at various temperatures. The intensity remains almost zero up to the transition field $H_N(T)$, and then increases rapidly. It is clear that field-induced Néel ordering takes place at $H_N(T)$. At 0.2 K, the intensity $(m_\perp^2)$ is almost proportional to $H - H_g$ just above H_g, and is slightly concave at higher fields. With increasing temperature, the transition field $H_N(T)$ increases, and the slope just above $H_N(T)$ increases.

The phase transition points obtained by the present measurements are summarized in Fig.4, where the data obtained from the magnetic measurements[3,8] and the specific heat measurements[9] are also plotted. Since the transition points determined from the present neutron scattering experiment and the other measurements lie almost on the same line, they are consistent with each other. The phase boundary can be described by the power law

$$[H_N(T) - H_g] \propto T^{\phi}\ , \qquad (2)$$

where $H_N(T)$ is the transition field at temperature T. We reevaluate the value of ϕ using the data for $T < 4$ K, which is lower than half the gap temperature $\Delta/k_B = 7.5$ K. The best fitting is obtained with $H_g = 5.63$ T and $\phi = 2.1(1)$. The exponent obtained, $\phi = 2.1(1)$, is somewhat larger than the $\phi = 1.5$ predicted by the magnon BEC theory based on the Hartree-Fock (HF) approximation. The difference between these values may be attributed to the fluctuation effect which is disregarded in the HF approximation.

§4. Conclusions

The field-induced transverse Néel ordering was clearly observed for $H > H_g \approx 5.6$ T in $TlCuCl_3$. The spin structure in the ordered phase was determined as shown in Fig. 1. The phase boundary can be represented by the power law, as predicted by the magnon BEC theory. However, the exponent obtained, $\phi = 2.1$, is somewhat larger than the predicted value of $\phi = 1.5$.

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