Bose-Einstein Condensation of Dilute Magnons in $TlCuCl_3$

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The recent observation [A. Oosawa *et al.,* J. Phys. Condens. Matter **11**, 265 (1999)] of the field-induced Néel ordering in the spin-gap magnetic compound $TlCuCl_3$ is interpreted as a Bose-Einstein condensation of magnons. A Hartree–Fock-type calculation based on this picture is shown to describe the temperature dependence of the magnetization well.

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The Bose-Einstein condensation (BEC) is one of the most exotic phenomena predicted by quantum mechanics. Recently, there has been a renewed interest in BEC, because the realization of BEC by ultracooling of dilute atoms has become possible [1]. While the BEC of ultracooled atoms is of great interest, there are various experimental limitations. On the other hand, it has been known for a long time that a quantum spin system can be mapped onto an interacting Bose gas, and that the off-diagonal long-range order which characterizes BEC corresponds to long-range magnetic order in the spin system [2]. It is then possible to tune the density of bosons (magnons) by a magnetic field to observe BEC of dilute bosons. However, an experimental realization of this BEC has not been reported so far.

In this Letter, we argue that BEC of dilute bosons in a thermodynamic number $\sim 10^{20}$ is realized in a recent high-field experiment on $TlCuCl_3$ [3], which is composed of a chemical double chain of Cu_2Cl_6 [3,4]. The compound has an excitation gap $\Delta/k_B \approx 7.5$ K above the singlet ground state, in the absence of the magnetic field [3,5,6]. The origin of the gap may be attributed to the antiferromagnetic dimer coupling in the double chain. When the external field $H_g = \Delta/(g\mu_B)$ to the gap is applied, the gap collapses. At finite temperature, the "collapse" of the gap at H_g does not give a singularity because thermal excitations exist even if $H < H_g$. However, there seems to be a phase transition due to the interchain interactions at higher field $H = H_c > H_g$, which depends on the temperature. In Ref. [3] the phase transition was identified as a long-range magnetic ordering, and was compared with a mean-field theory (MFT) [7] based on a dimer model. While the dimer MFT does predict the field-induced ordering, the experimental features were not well reproduced. In particular, it predicts almost flat dependence of the critical temperature T_c on the magnetic field, while in the experiment T_c depends on the magnetic field by a power law $T_c^{\phi} \sim H - H_g$ (see Fig. 1). Moreover, it predicts almost constant magnetization for $T < T_c$ and concave magnetization for $T > T_c$, as a function of temperature T. However, in the experiment, magnetization was found to increase as decreasing T below T_c and it is a convex function of T for $T > T_c$ (see Fig. 2).

We will show that the transition is rather well described as the BEC of magnons. While the details of the exchange interactions in $TlCuCl_3$ are not known yet, excitations above the singlet ground state generally can be treated as a collection of bosonic particles—magnons [8]. As discussed in Ref. [3], the magnetic anisotropy in $TlCuCl_3$ is negligible, in which case the number of magnons are conserved in a short time scale (but not conserved in a longer time scale). We assume that magnons carry spin 1, as generally expected.

Under a magnetic field $H \sim H_g$, the magnons with $S^z = 1$ can be created by small energy. Thus, at low temperatures $T \ll \Delta$ and $H \sim H_g$, we must consider only those magnons. The chemical potential of the magnons is given by $\mu = g\mu_B(H - H_g)$. The total number of magnons N is associated with the total magnetization M through $M = g\mu_B N$. If the magnons are free bosons, the number of magnons would be infinite for $H > H_g$. However, in the spin system, it is actually bounded due to a hard-core-type interaction between magnons.

The transverse components of the exchange interactions give rise to hopping of the magnons, while the longitudinal components give rise to the interaction. Although the

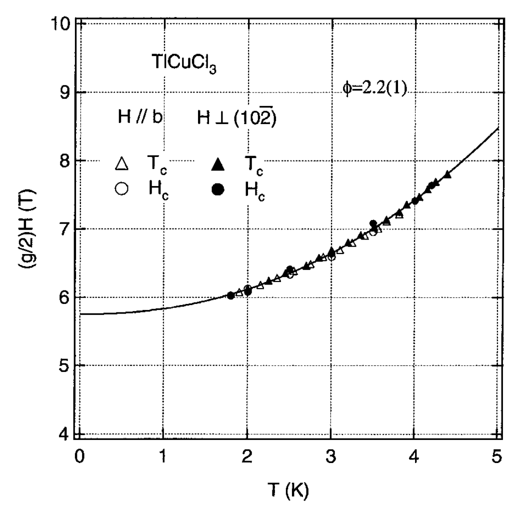

FIG. 1. The phase diagram in $TlCuCl_3$. The solid line denotes the fitting with the formula $(g/2)[H_c(T) - H_c(0)] \propto T^{\phi}$ with $(g/2)H_c(0) = 5.61$ T and $\phi = 2.2$.

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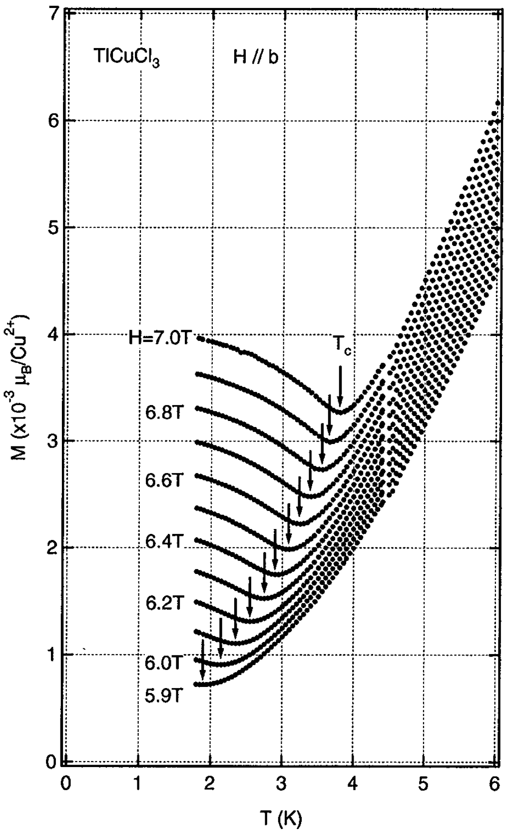

FIG. 2. The low-temperature magnetizations of $TlCuCl_3$ measured at various external fields for $H \parallel b$.

exchange interaction and thus hopping might be complicated, generically the dispersion relation of a magnon is quadratic near the bottom. Thus the low-energy effective Hamiltonian for the ($S^z = 1$) magnons is given by

$$H \sim \sum_k \left[\left(\sum_{\alpha=x,y,z} \frac{\hbar^2 k_\alpha^2}{2m_\alpha} \right) - \mu \right] a_k^\dagger a_k + \frac{1}{2} \sum_{k,k',q} v(\mathbf{q}) a_{k+q}^\dagger a_{k'-q}^\dagger a_k a_{k'} + \dots . \quad (1)$$

Here the momentum **k** is measured from the minimum of the magnon dispersion. For simplicity, we do not consider the case where the magnon dispersion has more than one minimum [9]. The effective masses m_α is related to the curvature of the dispersion relation in the direction of α. By a rescaling of momentum, we may consider isotropic effective Hamiltonian instead. This is nothing but the Hamiltonian for the nonrelativistic bosons with a short-range interaction.

Moreover, in the low-density and low-temperature limit, only the two-particle interaction is important and it can be replaced by delta-function interaction $v(q) \sim v_0$. Thus the effective Hamiltonian is given by

$$H = \sum_k \left(\frac{\hbar^2 k^2}{2m} - \mu \right) a_k^\dagger a_k + \frac{v_0}{2} \sum_{k,k',q} a_{k+q}^\dagger a_{k'-q}^\dagger a_k a_{k'} . \quad (2)$$

While this can be derived from some specific models [10,11], it is universal in the low-temperature and low magnon density limit, irrespective of the details of the exchange interaction.

Since the number of magnons is actually not conserved due to the small effects neglected in the Hamiltonian, we have a grand canonical ensemble of the bosons. The "chemical potential" can be controlled precisely by tuning the magnetic field. When the chemical potential becomes larger than a critical value, the system undergoes a BEC. Thus the spin-gap system in general would provide a great opportunity to study BEC in a grand canonical ensemble, with a thermodynamically large number of particles.

The idea that BEC is induced by the magnetic field in a spin-gap system has been discussed several times, for example, in Ref. [12] for a Haldane gap system. Giamarchi and Tsvelik [11] have recently discussed the three-dimensional ordering in coupled ladders in connection with BEC. However, as far as we know, there has been no experimental observation of magnon BEC induced by an applied field.

We first consider the normal (noncondensed) phase. Within the Hartree-Fock (HF) approximation, the momentum distribution of the magnons is given by [13]

$$n_k \equiv \langle a_k^\dagger a_k \rangle = \frac{1}{e^{\beta(\varepsilon_k - \mu_{\mathrm{eff}})} - 1} , \quad (3)$$

with $\varepsilon_k \equiv \hbar^2 k^2 / 2m$ and $\mu_{\mathrm{eff}} \equiv \mu - 2v_0 n$. The magnon density $n \equiv N/N_d$ (N_d is the total number of the dimer pairs) has to be determined self-consistently by

$$n = \int \frac{d^3k}{(2\pi)^3} n_k = \frac{1}{\Lambda^3} g_{3/2}(z) , \quad (4)$$

where $z \equiv e^{\beta\mu_{\mathrm{eff}}}$ is the fugacity, $\Lambda \equiv (2\pi\hbar^2/mk_\mathrm{B}T)^{1/2}$ is the thermal de Broglie wavelength, and $g_n(z) \equiv \sum_{l=1}^{\infty} z^l/l^n$ is the Bose-Einstein function. BEC occurs when the effective chemical potential μ_{eff} vanishes so that $\mu = 2v_0 n$. Setting $z = 1$ in (4) gives the temperature dependence of the critical value of the chemical potential

$$\mu_c = 2v_0 \left(\frac{mk_\mathrm{B}T}{2\pi\hbar^2} \right)^{3/2} \zeta(3/2) . \quad (5)$$

This implies that the temperature dependence of the critical magnetic field at low temperatures is $[H_c(T) - H_g] \propto T^{3/2}$. This power-law dependence is independent of the interaction parameter v_0.

When $\mu > \mu_c$, one has the macroscopic condensate order parameter $\langle a_0 \rangle = \sqrt{N_c}\, e^{i\theta} \neq 0$, where N_c is the total number of the condensate magnons. In terms of the original spin system, this means that there is a (staggered) transverse magnetization component $m_\perp = g\mu_\mathrm{B}\sqrt{n_c/2}$ with $n_c \equiv N_c/N_d$. Within the Hartree-Fock-Popov (HFP) approximation, the condensate density is determined by [14]

$$\mu = v_0 n_c + 2v_0 \tilde{n} , \quad (6)$$

where $\tilde{n} = n - n_c$ is the density of the noncondensed magnons, which is given by

$$\tilde{n} = \int \frac{d^3k}{(2\pi)^3}\left[\left(\frac{\varepsilon_k + v_0 n_c}{2E_k} - \frac{1}{2}\right) + \frac{\varepsilon_k + v_0 n_c}{E_k} f_{\rm B}(E_k)\right] = \frac{1}{3\pi^2}\left(\frac{m v_0 n_c}{\hbar^2}\right)^{3/2} + \int \frac{d^3k}{(2\pi)^3}\frac{\varepsilon_k + v_0 n_c}{E_k} f_{\rm B}(E_k), \quad (7)$$

where we have used the HFP energy spectrum $E_k = \sqrt{\varepsilon_k^2 + 2\varepsilon_k v_0 n_c}$ and the Bose distribution $f_{\rm B}(E_k) = 1/(e^{\beta E_k} - 1)$. The first term of (7) represents the depletion of the condensate due to interaction between magnons, which reduces to the ground-state noncondensate density at $T \to 0$. The second term is the contribution from thermally excited noncondensate magnons, which vanishes at $T \to 0$. Equation (6) is to be solved self-consistently in conjunction with Eq. (7). Then the total magnon density is given by

$$n = n_c + \tilde{n} = \frac{\mu}{v_0} - \tilde{n}. \quad (8)$$

In particular, the magnon density at $T \to 0$ is given by

$$n \approx \frac{\mu}{v_0} - \frac{1}{3\pi^2}\left(\frac{m\mu}{\hbar^2}\right)^{3/2}. \quad (9)$$

A simple approximation that ignores the deviation of z from 1 for $T \neq T_c$ is often used in the literatures [11,15]. As we will show later, while it is useful in obtaining an intuitive understanding, it is not justified. From Eqs. (4) and (8), the approximation $z = 1$ gives [we use $v_0 n_c = 0$ in (7) for $T < T_c$] [11]

$$\frac{n(T)}{n(T_c)} = \left(\frac{T}{T_c}\right)^{3/2} \quad (T > T_c), \qquad \frac{n(T)}{n(T_c)} = 2 - \left(\frac{T}{T_c}\right)^{3/2} \quad (T < T_c). \quad (10)$$

It predicts the cusplike minimum of the magnon density (magnetization) at $T = T_c$. In contrast, the dimer MFT [7] predicts a constant magnetization below T_c.

Figure 2 shows the observed low-temperature magnetizations of $\mathrm{TlCuCl_3}$ at various external fields for $H \parallel b$. We can see the cusplike anomaly at the transition temperature, as predicted by the present theory. The similar temperature dependence of the magnetization can be observed for $H \perp (1, 0, \bar{2})$ [3]. Our magnon BEC picture captures the main qualitative feature of the temperature dependence of the magnetization, which cannot be understood in the dimer MFT. The increase of n for decreasing T below T_c is due to condensation of the bosons; the cusp shape of the magnetization curve observed in the experiment can be regarded as evidence of the magnon BEC. We note that, in the range of the experiment, the magnon density is of order of 10^{-3} and is consistent with the assumption of diluteness.

However, the approximation (10) does not precisely reproduce the experimental result. In particular, it predicts independence of n on the applied field μ for $T > T_c$ while the dependence was observed experimentally. Part of the discrepancy may be due to the approximation $z = 1$. Actually, even in the HF framework, the approximation $z \sim 1$ is not really justified. In Fig. 3, we plot the temperature dependence of the total density n above and below the transition temperature T_c obtained by solving the self-consistency equations (4) and (8) numerically, without assuming $z = 1$. The interaction parameter v_0 and the effective mass m are estimated from the experimental data as $v_0/k_{\rm B} \approx 400$ K and $mk_{\rm B}/\hbar^2 \approx 0.025\ \mathrm{K^{-1}}$. The self-consistent calculation does predict that the total density n is dependent on the applied field for $T > T_c$, which is qualitatively consistent with the experiment. In Fig. 4 we also plot the temperature dependence of the staggered transverse magnetization component $m_\perp$. Direct measurements of $m_\perp$ using neutron diffraction are in progress.

We see a discontinuity in magnon density (magnetization) arises at the transition point in Fig. 3. This is because our HFP approximation is inappropriate in the critical region, and leads to an unphysical jump in the condensate density n_c [14]. In the vicinity of the critical point, the HFP approximation eventually breaks down; the critical behavior then belongs to the so-called 3D XY universality class [16]. On this ground, in the vicinity of T_c, the transverse magnetization $m_\perp$ is expected as $m_\perp \propto (T_c - T)^\beta$, where $\beta \sim 0.35$.

Figure 1 shows the experimentally determined magnetic phase diagram of $\mathrm{TlCuCl_3}$. We fit the phase boundary H_c as a function as a temperature T_c with the formula $(g/2) \times [H_c(T) - H_c(0)] \propto T^\phi$. The best fitting is obtained with $(g/2)H_c(0) = 5.61$ T and $\phi = 2.2$ [17]. The obtained exponent $\phi = 2.2$ disagrees somewhat with the HF approximation (5) which gives $\phi = 3/2$. We note that exactly $z = 1$ holds at the transition point, and thus $\phi = 3/2$ is a definite conclusion within the HF framework. On the other hand, the dimer MFT predicts $H_c(T)$ to be exponentially

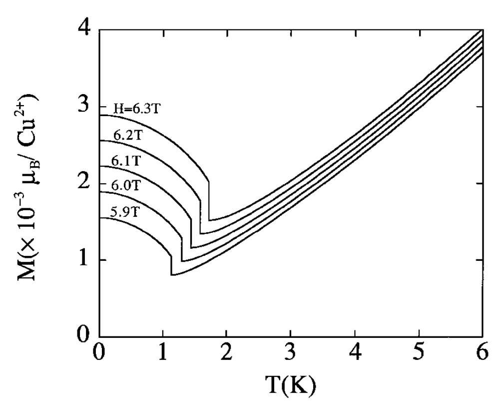

FIG. 3. The temperature dependence of the magnetization. We have used $v_0/k_{\rm B} = 400$ K, $mk_{\rm B}/\hbar^2 = 0.025\ \mathrm{K^{-1}}$, and $g = 2.06$.

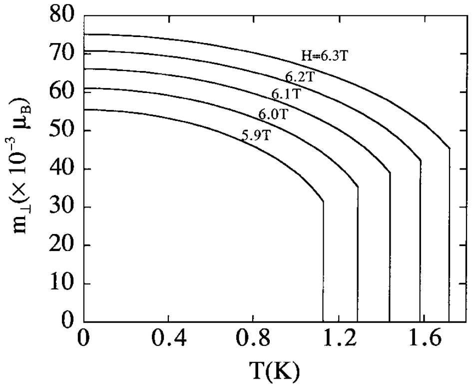

FIG. 4. The temperature dependence of the transverse magnetization $m_\perp$. We have used the same set of parameters as in Fig. 3.

flat at low temperature [7]. The observed power-law dependence is qualitatively consistent with the magnon BEC picture, compared to the dimer MFT.

On the other hand, a better description of the experimental data near the critical point requires more sophisticated analysis beyond the HFP approximation. Furthermore, in the experiment there may be other effects that were ignored in the effective Hamiltonian (2), such as impurities. These are interesting problems, to be studied in the future.

To conclude, the essential feature of the experimental observation on $TlCuCl_3$, which cannot be understood in the traditional dimer MFT, is captured by the magnon BEC picture. The present system could be the first experimental observation of a field-induced magnon BEC. It would give a new way of studying BEC in a grand canonical ensemble, with an easily tunable chemical potential (magnetic field) and a thermodynamically large number of particles. Similar BEC of magnons would be observed in various ladder compounds [18] which have a spin gap and spin-1 excitations. We also expect the magnon BEC in other magnetic systems in the vicinity of the gapped phase, which can be the singlet ground state due to large single-ion anisotropy [19], the completely polarized state [2,9], or the "plateau" phase in the middle of the magnetization curve [20]. An essential requirement for observing BEC is that the system is (approximately) rotationally invariant about the direction of the applied magnetic field, so that the number of magnons is conserved.

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