Bose-Einstein Condensation of Magnons in TlCuCl₃: Phase diagram and specific heat from a self-consistent Hartee-Fock calculation with a realistic dispersion relation.

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We extend the self-consistent Hartree-Fock-Popov calculations by Nikuni et al. [Phys. Rev. Lett. 84, 5868 (2000)] concerning the Bose-Einstein condensation of magnons in TlCuCl₃ to include a realistic dispersion of the excitations. The result for the critical field as a function of temperature behaves as $H_c(T) - H_c(0) \sim T^{3/2}$ below 2 K but deviates from this simple power-law at higher temperature and is in very good agreement with the experimental results. The specific heat is computed as a function of temperature for different values of the magnetic field. It shows a $\lambda$-like shape at the transition and is in good qualitative agreement with the results of Oosawa et al. [Phys. Rev. B 63, 134416 (2001)].

1. Introduction

TlCuCl₃ is a spin-$\frac{1}{2}$ magnetic insulator with a spin gap $\Delta = 7.5$ K.² It has been successfully described as copper dimers with an intra-dimer antiferromagnetic exchange energy $J \approx 5.5$ meV and weaker ($\lesssim 1.5$ meV) inter dimer couplings.³,⁴ At zero temperature, an applied magnetic field $H$ closes the gap at the critical field $H = H_c(0)$, giving rise to a quantum phase transition. $H_c(0)$ is related to the gap by $g\mu_B H_c(0) = \Delta$, where $\mu_B$ is the Bohr magneton and $g$ is the Lande $g$-factor. The field-induced phase transition continues to finite temperature $T$, with the temperature-dependent critical field $H_c(T)$. Above the critical field, a magnetic long-ranged order in the plane perpendicular to applied field develops.⁵,⁶ The existence of the ordering transition was predicted by a standard mean-field theory for spins.⁷ However, several characteristic features of the transition could not be explained by the mean-field theory. The two most notable features are the cusp-like minimum of the magnetization as a function of the temperature at the transition, and the power-law like dependence of the critical field

$$H_c(T) - H_c(0) \propto T^\phi$$

(1)

in the low temperature regime. The mean-field theory⁷ rather predicts a monotonic decrease of the magnetization and an exponentially fast approach of the critical field $H_c(T)$ to its zero-temperature limit $H_c(0)$, on lowering the temperature.

These features were successfully explained, at least qualitatively, as a Bose-Einstein condensation (BEC) of spin triplet excitations (magnons).⁸ The cusp-like minimum of the magnetization at the transition temperature is understood with the decrease of the non-condensed magnons at all temperatures and the increase of the condensed magnons below the transition, as the temperature is lowered. Moreover, the self-consistent Hartree-Fock-Popov (HFP) approximation on the magnon condensation gives the power-law dependence (1) with the exponent $\phi = 3/2$, if the dispersion of the magnons is taken to be quadratic.

As one can easily control the magnetic field, which corresponds to the chemical potential of the magnons, this system provides a new arena for the study of BEC, in a grand-canonical ensemble with a tunable chemical potential.⁹ However, the results of the HFP approximation given in Ref. 8 are not quite satisfactory to describe the experimental data in a quantitative manner. In order to further extend the study of magnon BEC, it would be important to improve the HFP approximation and clarify its range of validity.

One of the problems is that the HFP approximation predicts a discontinuous jump of the magnetization at the transition temperature, which is not observed. This is considered to be an artifact of the HFP approximation, and related to its breakdown due to strong fluctuation in the vicinity of the transition. In this paper we rather focus on another problem concerning the phase boundary. That is, while the experimental results are roughly in agreement with the power law (1), the reported values⁵,⁶,⁸,10,11 of the exponent $\phi = 1.67 \sim 2.2$ are consistently larger than the HFP prediction 3/2. Although it was suggested that the deviation is again due to the fluctuation effects, it has not been clarified.

In the present work, we extend the self-consistent HFP calculations⁸ by including a realistic dispersion calculated from microscopic models³,⁴ instead of the quadratic approximation $\epsilon_k \approx \frac{\Delta^2}{4m}$ used previously.⁸ The critical field $H_c(T)$ obtained by this method is in very good agreement with the experiments and represents a significant improvement over the simple quadratic approximation. Therefore the puzzle regarding the discrepancy of the exponent $\phi$ between the theory and the experiment is solved within the HFP framework. Here we note that there are related theoretical works¹²-¹⁵ on this problem. We will comment on them later in Discussions.

We also make several other checks of the HFP approximation with the experimental data, to show that HFP framework has a rather wide range of validity but the quadratic approximation fails above a rather low tem-
perature \sim 1 \text{ K} for TlCuCl₃. Finally, the specific heat is also computed and compared with the results of Oosawa et al.\textsuperscript{19}

2. Hamiltonian

As in Ref. 8, the Zeeman splitting is assumed to be sufficiently large compared to temperature so that only the singlet and the lowest triplet states of each dimer need to be considered. With this approximation the system is described by a hard-core boson Hamiltonian

\begin{equation}
\mathcal{H}_K = \sum_k \hat{b}_k^\dagger \hat{b}_k (\epsilon_k - \mu) + \mu_g (H - \Delta)
\end{equation}

where \( \epsilon_0 = 0 \). The magnon-magnon interactions are described by

\begin{equation}
\mathcal{H}_U = \frac{1}{2N} \sum_{q,k,k'} U_{q,k,k'} \hat{b}_k^\dagger \hat{b}_{k+q} \hat{b}_{k'-q}
\end{equation}

and we will neglect the \( q \)-dependence of \( U_q \) and set \( U_q = U \). As discussed by Nikuni et al.,\textsuperscript{8} this system undergoes a phase transition between a normal phase (at low field or high temperature) where the system is populated by thermally excited triplets to a “superfluid” phase where the bosons condense. This condensation is equivalent, in the spin language, to a field-induced three-dimensional magnetic ordering.

3. Hartree-Fock-Popov treatment of the condensed phase

We reproduce the Hartree-Fock-Popov (HFP) mean-field analysis of Eqs. 2-5 which was discussed in Ref. 8. For a strong enough magnetic field the zero-momentum state (we assume that \( H = \Delta + \tilde{\mathcal{H}}_K \)) and the external magnetic field \( H \):

\begin{equation}
\mathcal{H}_K = \sum_k \hat{b}_k^\dagger \hat{b}_k (\epsilon_k - \mu)
\end{equation}

\begin{equation}
\mathcal{H}_U = \frac{1}{2N} \sum_{q,k,k'} U_{q,k,k'} \hat{b}_k^\dagger \hat{b}_{k+q} \hat{b}_{k'-q}
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4. Dispersion relation for TlCuCl₃

The dispersion relation of triplet excitations in TlCuCl₃ was measured at \( T = 1.5 \text{ K} \) with inelastic neutrons scattering by Cavadinii et al.\textsuperscript{3} This dispersion relation was very well reproduced by Matsumoto et al.\textsuperscript{4} within a bond-operator formalism. Their result is:

\begin{equation}
\epsilon_k - a_k + \Delta_0 = \sqrt{(J + a_k)^2 - a_k^2}
\end{equation}

\begin{equation}
a_k = J \cos(k_x) + J_{a2c} \cos(2k_x + k_z) + 2J_{abc} \cos(k_x + k_z/2) \cos(k_y/2)
\end{equation}

\begin{equation}
J = 5.501 \text{ meV}, \quad J_a = -0.215 \text{ meV}
\end{equation}
Table I. Estimations of the gap (or critical field at zero temperature) from experiments. Mag. stands for magnetization, INS for inelastic neutron scattering, ESR for electron spin resonance, ENS for elastic neutron scattering (observation of the magnetic ordering) and $C_v$ for specific heat measurements.

| Ref.          | $\Delta_0$ (K) | Method          |
|---------------|----------------|-----------------|
| Shiramura et al.$^2$ (1997) | 7.5 | Mag.            |
| Tanaka et al.$^{16}$ (1998) | 7.68 | ESR            |
| Oosawa et al.$^7$ (1999) | 7.54 | Mag.            |
| Tanaka et al.$^5$ (2001) | 7.66 | ENS            |
| Cavadinì et al.$^3$ (2001) | 9.28 | INS            |
| Oosawa et al.$^{17}$ (2001) | 7.66 | $C_v$          |
| Oosawa et al.$^{19}$ (2002) | 7.54 | INS            |
| Ruegg et al.$^{20}$ (2003) | 8.2 | INS            |
| Shindo et al.$^{11}$ (2003) | 7.33 | $C_v$          |

$$J_{a2c} = -1.581 \text{ meV}, \ J_{abc} = 0.455 \text{ meV(25)}$$

where the Brillouin zone is doubled in the $z$ direction ($-2\pi \leq k_z < 2\pi$) to represent the two magnon branches. The momentum shift by $k_0 = (0, 0, 2\pi)$ just insures the consistency between our convention that $\epsilon_0 = 0$ and the location of the minimum of the dispersion at $k_0$ in Refs. 3, 4. The dispersion relation above has a gap of 0.7 meV, which is in agreement with the result of Ref. 3. However the studies based on a determination critical field as a function of temperature (see Table I) provide slightly smaller estimates for the gap ($\Delta_0 \sim 0.65$ meV) in TICuCl$_3$. Therefore we corrected the value of $J$ so that the dispersion relation is consistent with these data. The corrected value was chosen to insure $\Delta_0 = 0.65$ meV (or equivalently $(g/2)H_c(0) = 5.61T$):

$$J = 5.489 \text{ meV} \quad (26)$$

From the computation of curvature of $\epsilon_k$ around $k = 0$ the effective inverse mass $1/m$ is 43.66 K (in units where $\hbar^2/k_B = 1$), in agreement with the value taken in Ref. 8. Fig. 1 shows the experimental data of Cavadinì et al. with the $\epsilon_k$ given by Eqs. 22-26. The dotted line corresponds to the quadratic approximation; it only matches the full expression at very low energy.

5. Critical density

Within the HFP approximation the boson density $n_{cr}$ (or magnetization) at the transition is independent of the strength $U$ of the magnon-magnon interaction as well as independent of the value of the zero-field gap $\Delta_0$. It is obtained by setting $\mu^\text{eff} = 0$ in Eq. 15. If the full dispersion relation is used, the result has no adjustable parameter left. The result is shown Fig. 2 and is in good agreement with the experimental data. We note however that the discrepancy is larger when the field is applied along the $b$ direction. We do not know the reason of the discrepancy at present.

In the low-temperature limit, the quadratic approximation would become asymptotically exact within the HFP theory, giving$^{8, 22}$

$$n_{cr}(T \to 0) = \frac{1}{2} \xi_3/2 \left( \frac{T_m}{2\pi} \right)^{3/2} \quad (27)$$

However, this $\sim T^{3/2}$ behavior (dotted line in Fig. 2) is only recovered at very low temperature and $n_{cr}(T)$ shows significant deviations from Eq. 27 already at 2 K.

6. Critical field and interaction parameter $U$

In the HFP approximation the critical field $H_c(T)$ is related to the critical density by

$$\left( g/2 \right) [H_c(T) - H_c(0)] = 2Un_{cr}(T) \quad (28)$$
A linear relation between $H_c(T)$ and $n_{cr}(T)$ is indeed observed in the experimental data, as can be seen in Fig. 3. The least-square fits are performed in the low-density region (or equivalently low-temperature). The values obtained for $H_c(0)$ are in good agreement with most of the previous estimates (see Tab. I). These fits also provide an estimate for $U$ around 340 K. However, as can be seen in Fig. 4, a slightly smaller value for $U$ (320 K) gives a critical field $H_c(T)$ which is in very good agreement with all the available experimental data, even at high temperatures. This value is close to that obtained from a similar HFP analysis (including a small magnetic exchange anisotropy) of the magnetization curves.\textsuperscript{15}

In the literature the experimental data for $H_c(T)$ have been analyzed by fitting to the power-law (1). Values from $\phi = 1.67$ to 2.2 have been reported\textsuperscript{5,6,8,10,11} and it has been suggested that the deviation from the HFP theory ($\phi = 1.5$) could be caused by fluctuations effects beyond the mean-field approximation. From our results it appears that a realistic dispersion relation\textsuperscript{4} combined with an HFP treatment is able to reproduce the data accurately with a single adjustable parameter ($U$). It covers a wide temperature range from the very low temperature regime $\sim 1$ K where the quadratic approximation holds, up to $\sim 8$ K.

7. Specific heat

The specific heat of TlCuCl$_3$ under magnetic field was measured by Oosawa\textit{ et al.}\textsuperscript{10} and shows a peak (with an asymmetric $\lambda$ shape) at the transition. In this section we compare these results with the prediction of the HFP theory.

From Eq. 12 the expectation value of the energy per site in the normal phase ($n_c = 0$) is

$$\langle E \rangle = -Un^2 + \frac{1}{T} \int \frac{d^3k}{(2\pi)^3} \epsilon_k f_B(\epsilon_k, T)$$

(29)

The specific heat is obtained by differentiation with respect to temperature and we get:

$$C_v = \frac{1}{T} \int \frac{d^3k}{(2\pi)^3} \epsilon_k^2 \left( -\frac{\partial f_B}{\partial \epsilon_k} \right) + 2U \frac{\partial n}{\partial T} \int \frac{d^3k}{(2\pi)^3} \epsilon_k \frac{\partial f_B}{\partial \epsilon_k}$$

(30)

with $k_B = 1$ and

$$\frac{\partial n}{\partial T} = \frac{1}{1 - 2U} \int \frac{d^3k}{(2\pi)^3} \frac{\partial f_B}{\partial \epsilon_k}$$

(31)

In the condensed phase, Eq. 12 gives

$$\langle E \rangle = \int \frac{d^3k}{(2\pi)^3} E_k \left[ f_B(\epsilon_k, T) + \frac{1}{2} \right] - \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} \epsilon_k + C$$

(32)

After some algebra, we obtain the specific heat as:

$$C_v = \frac{1}{T} \int \frac{d^3k}{(2\pi)^3} E_k^2 \left( -\frac{\partial f_B}{\partial E} \right) + 2U \frac{\partial n}{\partial T} \left[ n_c - n - \frac{1}{2} \right]$$

$$+ \int \frac{d^3k}{(2\pi)^3} \epsilon_k \left( f_B(E) + \frac{1}{2} + E \frac{\partial f_B}{\partial E} \right)$$

(33)
Fig. 5. Specific heat (per dimer) under an applied field (along the \( b \) axis) minus the specific heat in zero field. Full lines: HFP results with \( U = 320K \). Circles: Measurements by Oosawa et al.\(^\text{10}^\) The results for the different values of \( H \) have been shifted by 0.04 for clarity.

with

\[
\frac{\partial n}{\partial T} = \frac{1}{1 - 2U} \int \frac{d^3k}{(2\pi)^3} \left( \tilde{\xi} \frac{\partial f_B}{\partial E} + \frac{\mu_{\text{eff}}}{E} (f_B + \frac{1}{2}) \right)
\]

In Figs. 5 and 6 the HFP results above are compared with the data of Oosawa et al. for two magnetic field orientations. The theoretical curves reproduce qualitatively the \( \lambda \) shape observed experimentally, although the height of the peak seems to be overestimated.

8. Discussions

In this paper, we have shown that taking the realistic dispersion relation determined from the microscopic theory and from the neutron scattering data, we can significantly improve the HFP approximation to explain the experimental data, especially the phase boundary curve \( H_c(T) \). It is now evident that, in TiCuCl\(_3\) the magnon dispersion curve is rather “steep” so that the quadratic approximation fails above a rather low temperature \( \sim 1 \) K.

It may be rather surprising that the HFP approximation, which is generally believed to fail in the critical region, describes a wide range of experimental data precisely. This appears to be the case, even though the HFP approximation still contains unsatisfactory features of predicting discontinuities in the magnetization and in the specific heat at the transition. These discontinuities are considered to be an artifact of the HFP approximation. The true behavior of the magnetization in the model (2) is believed to be continuous and that of the specific heat to show a sharp cusp (negative exponent \( \alpha \), see Ref. 23) at the transition, which is classified as the 3-dimensional XY universality class.

However, in fact, the experimental data on TiCuCl\(_3\) discussed in Section 7 does not show such a sharp singularity and is rather similar to the HFP prediction. This may be explained by small anisotropies (breaking the \( U(1) \) symmetry around the magnetic field direction), which are expected to exist in any real magnetic system. The fact that the observed moment in the ordered (condensed) phase of TiCuCl\(_3\) points to a constant direction\(^6\) suggest the presence of the anisotropy. Moreover, recently it is argued that a high-precision ESR measurement reveals the anisotropy.\(^24\) Such anisotropies induce a small gap and should reduce the thermal fluctuations (and thus the specific heat) in the vicinity of the transition, which could be also smeared out into a crossover. Since the breakdown of the HFP approximation is generally due to the critical fluctuation, the reduction of the critical region caused by the magnetic anisotropies may actually make the agreement with the HFP predictions better, although we did not take any anisotropy into our calculation. Recently, an HFP calculation including a (small) magnetic anisotropy was carried out by Sirker et al.\(^15\) and provided an improved description of the magnetization curves compared to that obtained.
from the isotropic model. They also emphasized that the HFP approximation should be valid outside a narrow critical regime.

Magnetic anisotropies are not the only corrections that may be added to the present model. Indeed, NMR measurements revealed that the transition to the ordered phase is (weakly) first order and accompanied by a simultaneous lattice distortion\(^{25}\) (see also Ref. 12). Spin-phonon interactions therefore seem to reduce the importance of critical fluctuations close to the transition while the remaining lattice distortion certainly induces some change in the magnetic field.\(^{26}\) An analysis of the consequences of such a magneto-elastic coupling is an interesting issue for further studies.

Finally, let us comment on related theoretical works. Sherman et al.\(^{12}\) discussed that the agreement of the HFP result to the experiment is better if the “relativistic” form \(\epsilon_k + \Delta \sim \sqrt{\epsilon_k^2 + \Delta^2}\) is assumed for the magnon dispersion relation.\(^{12}\) Our approach in this paper of modifying the dispersion is actually the same to theirs. However, we see no particular reason why we should take the relativistic form, although it may be a better approximation for TiCuCl\(_3\) than the quadratic one. In any case, ours would give a further improvement over Ref. 12 within the HFP framework.

In Refs. 13, 14, the phase boundary \(H_c(T)\) is studied numerically by a Monte Carlo method, for a dimer system on a cubic lattice. The result should contain effects from both the deviation of the dispersion from simple quadratic, and the fluctuation beyond HFP. While we cannot directly compare their result to ours as we deal with different models, the qualitative behavior is similar. Namely, they also observed the deviation from \(\phi = 3/2\) at higher temperatures, but the result seems to become closer to the \(\phi = 3/2\) as the temperature is lowered. However, they suggest that this behavior including the deviation from \(\phi = 3/2\) could be universal and does not depend on the particular dispersion, in a moderately low temperature regime. This is in contrast to our result that the non-universal magnon dispersion explains the observed phase boundary \(H_c(T)\) and its deviation from \(\phi = 3/2\). The resolution is an open problem for the future. Numerical approaches would be also useful to clarify the effect of the (small) anisotropies.

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Note Added

After submission of this paper, Kawashima\(^{27}\) clarified the question of the exponent \(\phi\) with numerical simulations of the 3D S=1/2 XXZ model as well as field-theoretical arguments. According to his results, in the limit of \(T \to 0\), the HFP prediction \(\phi = 3/2\) is indeed exact. This is also consistent with our result that the phase boundary for a wide temperature range can be accounted within the HFP calculation using the realistic dispersion curve.

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