High-field instability of field-induced triplon Bose-Einstein condensate

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Abstract

We study properties of magnetic field-induced Bose-Einstein condensate of triplons as a function of temperature and the field within the Hartree-Fock-Bogoliubov approach including the anomalous density. We show that the magnetization is continuous across the transition, in agreement with the experiment. In sufficiently strong fields the condensate becomes unstable due to triplon-triplon repulsion. As a result, the system is characterized by two critical magnetic fields: one producing the condensate and the other destroying it. We show that nonparabolic triplon dispersion arising due to the gapped bare spectrum and the crystal structure has a strong influence on the phase diagram.

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Bose-Einstein condensation (BEC), a macroscopic quantum phenomenon, occurs in various systems of bosons, including, in addition to atoms, quasiparticles in systems out of equilibrium such as excitons and polaritons (for example,[1]). Theory predicts that quantum spin excitations in solids, being Bose-quasiparticles, can at certain conditions build the condensate, and magnetic ordering in various systems can be understood in terms of the BEC of these excitations.[2–6] The first experimental observation [7] of magnetic field-induced BEC of triplons, that is the spin $S = 1$ quasiparticles, in antiferromagnetic TlCuCl$_3$ produced a diverse research field. [8–17] In this compound, the triplon branches with $S_z = -1, 0, 1$, are separated from the ground state by a relatively small gap $\Delta$. For this reason, the Zeeman interaction in a modest external magnetic field $H_{\text{ext}}$ can close the gap for the $S_z = -1$ states. In contrast to atomic gases, where the total particle number is constant, for triplons it is proportional to magnetization $M(T, H_{\text{ext}})$ induced by $H_{\text{ext}}$. The density of triplons rapidly increases with the field, and they undergo the BEC leading to a magnetic ordering. This field-induced BEC, which occurs at the scale on the order of few K, has been observed in a variety of quantum antiferromagnets.[17]

The condensate properties crucially depend on interaction of the particles.[18] For the atomic BEC at $T = 0$ the interatomic repulsion can lead to the condensate instability when the concentration becomes large enough.[19] Another general feature clearly seen in the triplon BEC is the dependence of its physics on the bare dispersion of the quasiparticles $\varepsilon_k$. The non-parabolic bare dispersion of triplons [20] leads to a non-trivial dependence of the transition temperature $T_c$ on the concentration $\rho \sim M(T, H_{\text{ext}})$ and, hence, on $H_{\text{ext}}$. The bare dispersion, being itself $H_{\text{ext}}$-independent, determines the interplay of kinetic and potential energy of a macroscopic system, and, therefore, plays a crucial role in the BEC properties. The effects of the bare dispersion are clearly seen experimentally as the $\rho$-dependence $T_c \sim \rho^{\phi(\rho)}$. The exponent $\phi(\rho)$ approaches $2/3$ at low concentrations (low $T_c$),[11] as predicted for the parabolic $\varepsilon_k$, while at $T > 2.5$ K, $\phi(\rho)$ is close to 0.5.

Here we establish theoretical phase diagram of the field-induced triplon BEC based on the Hartree-Fock-Bogoliubov (HFB) approximation taking into account also a nonparabolic dispersion and determine the fields $H_{\text{ext}}^{(1)}$ and $H_{\text{ext}}^{(2)} > H_{\text{ext}}^{(1)}$, corresponding to the BEC onset and to the instability. A problem in the current theoretical description of the transition at $H_{\text{ext}}^{(1)}$ is its predicted discontinuity. We show that this result is an artefact of the conventional Hartree-Fock-Popov (HFP) approximation, neglecting the anomalous density terms. When
the anomalous density is taken into account, the theory correctly predicts the continuous transition. For this reason, the HFB method is more appropriate to study the BEC than the HFP one. We find here the stability region of the triplon BEC in the $T - H_{\text{ext}}$ plane and prove that its boundaries strongly depend on the dispersion $\varepsilon_k$. Results on triplons and on cold atoms can be compared to foster the understanding of the similarities and differences of their BEC.

The triplons form a non-ideal Bose gas [4, 7, 20] with contact repulsive interaction described by the Hamiltonian:

$$\hat{H} = \int dV \left\{ \psi^\dagger(r) \mathcal{K} \psi(r) + \frac{g}{2} [\psi^\dagger(r) \psi(r)]^2 \right\},$$

(1)

where $\mathcal{K}$ is the kinetic energy operator and $g$ is the coupling constant, and we adopt the units $k_B = 1$, $\hbar = 1$, and $V = 1$ for the crystal volume. Below the critical temperature $T_c$ the global gauge symmetry becomes broken as realized by the Bogoliubov shift in the field operator: $\psi(r) = v + \tilde{\psi}(r)$. Here the condensate order parameter $v$ and $\tilde{\psi}(r)$ define the density of condensed and uncondensed particles, respectively: $\rho_0 = v^2$, $\rho_1 = \langle \tilde{\psi}^\dagger(r) \tilde{\psi}(r) \rangle$.

The grand canonical Hamiltonian is: $\hat{H}_G = \hat{H} - \mu \rho$, where $\mu$ is the chemical potential and the total density $\rho = \rho_0 + \rho_1$ is uniquely determined by $H_{\text{ext}}$. The density $\rho$ is considered as a dimensionless quantity. After the Bogoliubov shift one presents the grand Hamiltonian $\hat{H}_G$ in terms of second quantization operators as $\hat{H}_G = H_0 + H_1 + H_2 + H_3 + H_4$ with:[21]

$$H_0 = -\mu \rho_0 + \frac{g \rho_0^2}{2},$$

$$H_2 = \sum_k' \left[ (\varepsilon_k - \mu + 2g\rho_0) a_k^\dagger a_k + \frac{g\rho_0}{2} (a_k a_{-k} + \text{h.c.}) \right],$$

$$H_4 = \frac{g}{2} \sum_{k,p,q} a_k^\dagger a_p^\dagger a_q a_{k+p-q},$$

where the prime shows that zero momentum states are excluded. Similarly defined linear ($H_1$) and cubic ($H_3$) terms having zero mean-field approximation (MFA) expectation values are omitted.

Now we implement the HFB approximation [21, 22]:

$$a_k^\dagger a_p^\dagger a_q a_m \rightarrow 4a_k^\dagger a_m (a_q^\dagger a_p) + a_q a_m (a_k^\dagger a_p^\dagger) + a_k^\dagger a_p^\dagger (a_q a_m) - 2\rho_1^2 - \sigma_1^2,$$

(3)

where $\langle a_k^\dagger a_p \rangle = \delta_{k,p} n_k$, $\langle a_k a_p \rangle = \delta_{k,-p} \sigma_k$, $n_k$ and $\sigma_k$ are related to the normal $\rho_1 = \sum_k n_k$ and anomalous $\sigma_1 = \sum_k \sigma_k$ densities. The grand Hamiltonian in this approximation involves
only zero $\tilde{H}_0$ and second order $\tilde{H}_2$ contributions in $a_k, a_k^\dagger$:

$$\tilde{H}_0 = -\mu\rho_0 + \frac{g}{2} \left[ \rho_0^2 - 2\rho_1^2 - \sigma_1^2 \right],$$
$$\tilde{H}_2 = \sum_k' \left[ \omega_k a_k^\dagger a_k^\dagger + \frac{X_1}{4} (a_k a_{-k} + \text{h.c.}) \right],$$

where $\omega_k = \varepsilon_k - \mu + 2\rho_0$ and

$$X_1 = 2g(\rho_0 + \sigma_1).$$

(4)

It follows from (4) that for $T > T_c$ the $\tilde{H}_2$ term is diagonal and hence, the triplon density is given by the same formula as in the widely used HFP approximation

$$\rho(T > T_c) = \sum_k \frac{1}{e^{\omega_k/T} - 1} = \sum_k \frac{1}{e^{(\omega_k - \mu_{\text{eff}})/T} - 1},$$

(6)

where $\mu_{\text{eff}} = \mu - 2\rho_0$. In the BEC regime one performs Bogoliubov transformation

$$a_k = u_k b_k + v_k b_{-k}^\dagger, \quad a_k^\dagger = u_k b_k^\dagger + v_k b_{-k},$$

(7)

with $[b_k, b_p^\dagger] = \delta_{k,p}$, $\langle b_k^\dagger b_{-k}^\dagger \rangle = \langle b_k b_{-k} \rangle = 0$. As a result, the grand Hamiltonian is transformed to the Bogoliubov form:

$$H = \tilde{H}_0 + \sum_k E_k b_k^\dagger b_k + \frac{1}{2} \sum_k (E_k - \omega_k),$$

(8)

where $\langle b_k^\dagger b_k \rangle = n_B(E_k, T) = 1/[\exp(E_k/T) - 1]$ with the phonon Goldstone mode dispersion $E_k^2 = \omega_k^2 - X_1^2/4$. At small momenta, this mode is a collective excitation of the condensate carrying spin $S_z = -1$, while at large momenta it becomes the bare triplon mode.

In accordance with Hugenholtz-Pines theorem [23] at small $k$ the phonon dispersion is linear: $E_k \sim c k + O(k^2)$, where $c$ can be interpreted as the speed of sound. This is achieved by setting $\omega_k - X_1/2 = \varepsilon_k$, that is, by:

$$\mu - g\rho_0 - 2g\rho_1 + g\sigma_1 = 0.$$

(9)

This choice yields $E_k = \sqrt{\varepsilon_k(\varepsilon_k + X_1)}$ with $c = \sqrt{X_1/2m}$, where $m$ is the triplon effective mass. It can be shown [19, 21] that $X_1$ is related to the normal and anomalous self energies as $\Sigma_n = X_1/2 + \mu$ and $\Sigma_a = X_1/2$, respectively. The quantity $X_1$ plays a special role in our analysis: when $X_1 > 0$, the condensate is stable, otherwise it decays due to triplon-triplon interaction. [24–26] Below we find $X_1$ in the $T - H_{\text{ext}}$ plane and determine the stable BEC region by the condition $X_1 \geq 0$. 

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Using the explicit \( u_k = \sqrt{(\omega_k + E_k)/2E_k} \) and \( v_k = \sqrt{(\omega_k - E_k)/2E_k} \), one obtains:

\[
\begin{align*}
\rho_1 &= \sum_k \langle a_k^+ a_k \rangle = \sum_k \left( \frac{\omega_k W_k}{E_k} - \frac{1}{2} \right), \\
\sigma_1 &= \sum_k \langle a_k a_{-k} \rangle = 2 \sum_k u_k v_k W_k = -\frac{X_1}{2} \sum_k \frac{W_k}{E_k},
\end{align*}
\]

(10)

where \( W_k = n_B(E_k, T) + 1/2 \). Near the transition, \( T \to T_c \) the condensate fraction vanishes: \( \rho_0 \to 0 \), and Eq.(5) yields \( X_1 = 0 \). In the triplon BEC, the critical density \( \rho_c \) corresponds to \( \mu_{\text{eff}} = 0 \), i.e. \( \rho_c = \mu/2g \). Therefore, at a given chemical potential \( \mu = \tilde{g}\mu_B H_{\text{ext}} - \Delta \), where \( \tilde{g} \) is the electron Landé-factor, \( T_c \) is determined by: \( \Sigma_k n_B(\varepsilon_k, T_c) = \mu/2g \).

To perform MFA calculations one starts by solving Eqs.(5) and (9) with \( \rho_1 \) and \( \sigma_1 \) given by Eq.(10). In contrast to the BEC of atomic gases, in the triplon problem, the chemical potential is the input parameter, whereas the densities are the output ones. Bearing this in mind, we rewrite Eqs.(5) and (9) as:

\[
\begin{align*}
X_1 &= 2\mu + 4g(\sigma_1 - \rho_1), \\
\rho_0 &= \frac{X_1}{2g} - \sigma_1.
\end{align*}
\]

(11)

Using dimensional regularization at \( T = 0 \), we can find from (10) more explicit expressions for the densities

\[
\begin{align*}
\rho_1 &= \rho_1(T = 0) + \int \frac{d^3k}{(2\pi)^3} n_B(E_k, T) \frac{\varepsilon_k + X_1/2}{E_k}, \\
\sigma_1 &= \sigma_1(T = 0) - \int \frac{d^3k}{(2\pi)^3} n_B(E_k, T) \frac{X_1/2}{E_k},
\end{align*}
\]

(12)

where \( \sigma_1(T = 0) = 3\rho_1(T = 0) = \sqrt{2}(mX_1)^{3/2}/4\pi^2 \), as shown in Ref.[22]. By setting in all above formulas \( \sigma_1 = 0 \), one arrives at the HFP approximation, [7, 20] and particularly

\[
\begin{align*}
X_1^{[\text{HFP}]} &= 2\mu - 4g\rho_1, \\
\rho_0 &= \frac{X_1^{[\text{HFP}]}_1}{2g}.
\end{align*}
\]

(13)

The above Eqs.(11)-(13) can be applied for any realistic \( \varepsilon_k \). It is instructive to note that for the parabolic dispersion \( \varepsilon_k = k^2/2m \), the BEC can be fully described by only two parameters \( \eta \equiv \mu m^3 g^2 \) and \( t \equiv T/T_c \) with \( T_c = \tilde{c}(\mu/g)^{2/3} / m, \tilde{c} = \pi \left( \sqrt{2}/g_{3/2}(1) \right)^{2/3} = 2.0867 \), where \( g_{3/2}(z) \) is the Bose function.[18] The parameter \( \eta \) is an analogue of the gas parameter [18] of atomic BEC.

Since the MFA (both HFB and HFP) calculations are based on Eqs.(11), (12), a question about the existence of positive solutions for \( X_1 \) arises. To analyze qualitatively the existence of the physical solutions, we consider \( T = 0 \) case. Here, the HFP Eq.(13) is simplified by
FIG. 1: (Color online). Comparison of the HFB (solid) and the HFP (dashed lines) results for the triplon density. The HFB approach shows a continuous behavior, which fully agrees with the experimental data [11, 12] while the HFP approach leads to the discontinuity. The corresponding $H_{\text{ext}}$ are marked near the plots.

Substitution $Z_{\text{HFP}} \equiv X_1^{[\text{HFP}]} / 2\mu$ to $1 = Z_{\text{HFP}} + 2Z_{\text{HFP}}^{3/2} \sqrt{\eta} / 3\pi^2$ and has physical solutions $Z_{\text{HFP}} > 0$ for any $\eta > 0$. This remains valid for all $t \leq 1$ at any concentration $\rho$. However, in the HFB approximation the situation is different: even at $t \leq 1$, the physical solutions of Eq.(11) can disappear if $\eta$ exceeds a critical value $\eta_c$. For example, at $T = 0$, Eq. (11) for $Z \equiv X_1 / 2\mu$ simplifies as

$$1 = Z - \frac{4Z^{3/2}\sqrt{\eta}}{3\pi^2}. \tag{14}$$

When $\eta$ exceeds $\eta_c = \pi^4 / 12$, the rhs in Eq.(14) is less than 1 for any $Z \geq 0$, therefore, it has no positive solutions, and, as a result, $X_1$ acquires an imaginary part. Bearing in mind that $\eta = \mu m^3 g^2 = (\mu B g H_{\text{ext}} - \Delta) m^3 g^2$, one concludes that even at $T = 0$, if the $H_{\text{ext}}$ is strong enough the speed of sound $c = \sqrt{Z\mu/m}$ becomes complex and, hence, the BEC is unstable.

To calculate $\rho$ and $X_1$ one needs the bare $\varepsilon_k$. Misguich and Oshikawa [20] demonstrated that only with the exact $\varepsilon_k$ one can explain the overall $\rho_c - T_c$ - dependence. Here we apply a similar approach, using a simpler, ”relativistic” $\varepsilon_k = \sqrt{\Delta^2 + J^2 k^2 / 4 - \Delta}$, generic for systems with gapped spectrum. This choice leads to $\rho_c \sim T_c^2$ at higher and $\rho_c \sim T_c^{3/2}$ at lower $T$s, respectively.[27, 28] Here the effective exchange $J = 2\sqrt{\Delta/m}$ is chosen to match the parabolic and the relativistic $\varepsilon_k$ at small $k$.

In numerical calculations we used parameters by Yamada et al. [11] for TlCuCl$_3$: $m = 0.0204$ K$^{-1}$, (i.e. $m = 0.261 \times 10^{-25}$ g), unit cell size 0.79 nm, $\Delta = 7.1$ K, $g = 313$ K and $\tilde{g} = 2.06$. We neglect a weak renormalization of the model parameters by temperature-
dependent many-body effects, which can slightly shift the stability region boundary, since we consider the regime of low $T$ and $\rho$. This assumption yields a perfect agreement of theory and experiment [20] in a similar range of $T$ and $\rho$. We begin with the comparison of the HFB and HFP approaches for the density $\rho$ in a constant $H_{\text{ext}}$. Fig.1 shows a continuous plot of $\rho(T, H_{\text{ext}})$ obtained with the HFB approach, [29] in full agreement with the experiment [11, 12] and in contrast to the HFP approximation. In Fig.2 we present the phase diagram obtained in the HFB for the parabolic and the relativistic $\varepsilon_k$. Solid curves in these figures present $T_c$ vs $H_{\text{ext}}$ obtained from $\Sigma_k n_{\uparrow}\varepsilon_k, T_c = \mu/2g$. The dashed lines present the BEC stability boundary: there is no $X_1 > 0$ solutions to the gap equations in the regions below these lines. Therefore, the HFB approach predicts the existence of a stable (the region between solid and dashed lines) and unstable BEC zones (the region below the dashed line). As expected, at low $T$ and small $H_{\text{ext}}$ the stability region in Figs. 2(a) and 2(b) is the same for both $\varepsilon_k$. In general, the relativistic dispersion leads to a narrower stability zone than the parabolic one. Note that magnetization measurements on TlCuCl$_3$ have been done for $H_{\text{ext}}$ between 5.1 and 9 T. [11, 12] It would be interesting to experimentally study its behavior at higher $H_{\text{ext}}$ to explore the instability region. [30] A direct access to the dispersion and damping of the phonon-like mode in TlCuCl$_3$ can be achieved in the inelastic neutron.
FIG. 3: (Color online). Triplon density as a function of $T$ for relativistic (solid) and parabolic (dashed lines) dispersion in the HFB approximation for $H_{\text{ext}}$ marked near the plots. The plot for $H = 12.5$ T (upper curves) shows two anomalies, one of them caused by the instability.

Density $\rho$ as a function of temperature is presented in Fig.3 for two $H_{\text{ext}}$. At relatively weak fields, e.g. $H_{\text{ext}} = 7.0$ T the magnetization exhibits only one anomaly at $T = T_c$ while at stronger one $H_{\text{ext}} = 12.5$ T, two anomalies are present. The minimum at the solid line at 6.2 K is the onset of the BEC, while the anomaly at $T$ slightly less than 3 K is due to the condensate decay. Similar physical behavior can be seen in Fig.4, which shows the BEC fraction $\rho_0/\rho \times 100\%$. This fraction is rather large ($\sim 95\%$ for $H_{\text{ext}} = 7.5$ T at $T = 0$) and rapidly decreases with increasing the temperature. In both Figs.3 and 4 the curves for $H_{\text{ext}} = 12.5$ T start at $T \approx 3$ K since the BEC is unstable below this $T$. However, Fig.4 shows that even close to this point the condensate fraction is approximately 70%, and, therefore, in the instability zone the condensate can exist for a short time [32] determined by the imaginary part of the self energy $\chi_1$. This regime will be considered in an extended paper.

In summary, we have theoretically established the phase diagram of the field-induced triplon BEC in quantum antiferromagnets in the $T - H_{\text{ext}}$ plane for a model relevant for the TlCuCl$_3$ compound. Our approach is based on the HFB approximation taking into account the anomalous density in the condensate phase. We have shown that (i) at the BEC transition the magnetization remains continuous demonstrating a minimum, in agreement with the experiment, (ii) in high magnetic fields the condensate becomes unstable due to the triplon-triplon repulsion, resulting in interaction of quasiparticles, and found the stability boundaries. The non-parabolic dispersion of triplons determined by the crystal structure.
FIG. 4: (Color online). Condensate density fraction for relativistic (solid line) and parabolic (dashed line) $\varepsilon_k$ in the HFB approximation for $H_{\text{ext}}$ marked near the plots.

has the crucial effect on the phase diagram by changing the boundaries $H_{\text{ext}}^{(1)}$ and $H_{\text{ext}}^{(2)}$ and making the stability region smaller.

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