Effects of exchange and weak Dzyaloshinsky–Moriya anisotropies on thermodynamic characteristics of spin-gapped magnets

Abdulla Rakhimov*†‡, Asliddin Khudoyberdiev*§ and B. Tanatar†¶

*Institute of Nuclear Physics, 100214 Tashkent, Uzbekistan
†Department of Physics, Bilkent University, Bilkent 06800, Ankara, Turkey
‡rakhimovabd@yandex.ru
§asliddinhk@gmail.com
¶tanatar@fen.bilkent.edu.tr

Received 30 March 2021
Revised 27 May 2021
Accepted 28 June 2021
Published 8 October 2021

We study the modification of low-temperature properties of quantum magnets such as magnetization, heat capacity, energy spectrum and densities of condensed and non-condensed quasiparticles (triplons) due to anisotropies in the framework of mean-field-based approach. We show that in contrast to exchange anisotropy (EA) interaction, Dzyaloshinsky–Moriya (DM) interaction modifies the physics dramatically. Particularly, it changes the sign of the anomalous density in the whole range of temperatures. Its critical behavior is slightly modified also by the EA. We have found that the shift of the critical temperature of phase transition (or crossover caused by DM interaction) is positive and significant. Using the experimental data on the magnetization of the compound TlCuCl$_3$, we have found optimal values for the strengths of EA and DM interactions. The spectrum of the energy of low lying excitations has also been investigated and found to develop a linear dispersion similar to Goldstone mode with a negligibly small anisotropy gap.

Keywords: Bose–Einstein condensation of triplons; exchange and Dzyaloshinsky–Moriya anisotropy; energy dispersion.

PACS numbers: 03.75.Kk, 75.30.Gw, 75.30.Et, 75.50.Ee

1. Introduction

Presently, it is well established that there is a class of quantum magnets whose low-temperature properties could be described within the paradigm of Bose–Einstein condensation (BEC) of quasiparticles referred as triplons.$^1$ Experimentally this is

$^1$Corresponding author.
confirmed by studying the critical exponents as well as the magnetic excitation spectrum of such compounds at low-temperatures. A good example is the critical exponent $\phi$, associated with phase boundary $H_c(T)$, that divides the paramagnetic and field-induced canted XY antiferromagnetic phase of several quantum magnets, $H_c(T) - H_c(0) \propto T^{\phi}$. This exponent approaches its expected value of 3/2, which is typical for a system with BEC, when the window of low-temperatures is rather reduced.\(^2\) Recent experimental investigations were conducted by Zhou et al.\(^3\) for the entire magnetization process of TlCuCl$_3$ up to the magnetic field of 100 T at temperature 2 K. They also analyzed magnetic field–temperature phase boundary dependence around the critical fields $H_{c1}$ and $H_{c2}$ and concluded that for both critical phase boundaries the critical exponents are $\phi \approx 3/2$. Another experimental evidence is offered by the properties of the excitation spectrum in the BEC state which has been theoretically predicted to be a gapless Goldstone mode associated with the spontaneous breaking of rotational symmetry by the staggered order. Thus, the presence of a spin-wave like mode with a linear mode dispersion, $E_k \sim c|k|$, is a convincing signal for the existence of BEC in this class of quantum magnets.\(^4\) Therefore, one may conclude that at low-temperatures thermodynamic properties of such materials are determined mainly (but not only) by the condensation of triplons.\(^5\)

Theoretically, the system of triplons can be described by the following effective Hamiltonian:

$$H_{iso} = \int d^3r \left[ \psi^+(\vec{r})(\vec{K} - \mu)\psi(\vec{r}) + \frac{U}{2}(\psi^+(\vec{r})\psi(\vec{r}))^2 \right],$$  

where $\psi(r)$ is the bosonic field operator, $\vec{K}$ is the kinetic energy operator which defines the bare triplon dispersion $\varepsilon_k$ in momentum space and $U$ is the strength of contact interaction describing a strong short-range triplon–triplon repulsion. The Hamiltonian in Eq. (1) is formally the same as used for BEC of atomic gases.\(^6\) However, there is a small difference in the strategy. In tasks related to atomic Bose gases, the number of particles $N$ is assumed to be fixed, while the chemical potential $\mu(N,T)$ is to be calculated say, by the relation $N \sim \sum_k 1/[e^{\beta(\varepsilon_k - \mu)} - 1]$, where $\beta$ is the inverse temperature. As to the triplon gas, the chemical potential in Eq. (1) characterizes an additional direct contribution to the triplon energy due to the external magnetic field $H$, giving $\mu = g\mu_B(H - H_c)$ where $g$ is the electron Landé factor, $\mu_B = 0.672$ KT$^{-1}$ is the Bohr magneton and $H_c$ is the critical magnetic field which defines the gap $\Delta_{ST} = g\mu_B H_c$ between singlet and triplet states. In the field-induced BEC, $\mu$ is assumed to be an input parameter, from which the total number of triplons can be calculated. Moreover, for homogenous atomic gases one may use simple quadratic bare dispersion $\varepsilon_k = k^2/2m$ with a good accuracy, while for spin-gapped quantum magnets a more complicated form of bare dispersion is needed.\(^2,7–10\)

It is well-known that the Hamiltonian in Eq. (1) leads to a gapless Bogoliubov dispersion, $E_k = \sqrt{\varepsilon_k^2 + 2U\rho} \approx ck + O(k^3)$ at low-temperatures,
Effects of exchange and weak DM anisotropies

with density $\rho = N/V$ and sound velocity $c$. However, low frequency electron spin resonance (ESR) measurements on some materials, such as TlCuCl$_3$, CS$_2$CuCl$_4$, DTN$^{15}$ gave evidence for a tiny spin gap. The origin of this gap is due to exchange anisotropy (EA) or Dzyaloshinsky–Moriya (DM) interactions, which should be taken into account in the theoretical description, and particularly, in the effective model Hamiltonian.$^{16}$ A simpler extended Hamiltonian such as Eq. (1) including EA and DM interactions was proposed by Sirker et al.$^{17}$

$$H_{\text{aniz}} = \int d^3r \{ \psi^+(\vec{r})(\hat{\mathbf{K}} - \mu)\psi(\vec{r}) + \frac{U}{2}(\psi^+(\vec{r})\psi(\vec{r}))^2 + \frac{\gamma}{2}[\psi^+(\vec{r})\psi^+(\vec{r}) + \psi(\vec{r})\psi(\vec{r})] + i\gamma'[\psi(\vec{r}) - \psi^+(\vec{r})] \}, \quad (2)$$

where $\gamma$ and $\gamma'$ are interaction strengths of EA and DM interactions, respectively ($\gamma \geq 0$, $\gamma' \geq 0$). Thus, once the Hamiltonian is given, one first separates fluctuations as $\psi = \xi\sqrt{\rho_0} + \tilde{\psi}$, where $\xi = e^{i\Theta}$ and $\rho_0$ are the phase of the condensate wavefunction and its magnitude, respectively; and then introducing second quantization, $\tilde{\psi}(\vec{r}) = \sum_k e^{ik\vec{r}\cdot\vec{r}}a_k$, $\tilde{\psi}^+(\vec{r}) = \sum_k e^{-ik\vec{r}\cdot\vec{r}}a_k^+$, makes an attempt to diagonalize the Hamiltonian $H$ with respect to creation $(a^+)$ and annihilation $(a)$ operators. As a result, analytical expressions for quasi-particle (bogolon) dispersion $E_k$ and some other quantities may be obtained. In the present work, we shall take into account anomalous averages $\sigma = \sum_k \sigma_k = \frac{1}{2} \sum_k \langle a_k a_{-k} \rangle = \frac{1}{2} \sum_k \langle a_k^+ a_{-k}^+ \rangle$ ($\sigma$-anomalous density) based on Hartree–Fock–Bogoliubov approach, which was neglected in Ref. 17. This allows one to obtain continuous magnetization across the BEC transition, which would be discontinuous otherwise, in the so-called Hartree–Fock–Popov (HFP) approximation with $\sigma = 0$.\footnote{In order to get more information about thermodynamics of the system, we exploit the grand canonical thermodynamic potential $\Omega$, which may be evaluated in the path integral formalism.$^{6,19–21}$ This will be convenient to study the modification of the condensate wavefunction, entropy $S = -(\partial\Omega/\partial T)$, heat capacity $C_H = T(\partial S/\partial T)$, magnetization $M = -(\partial\Omega/\partial H)$, and possibly other physical quantities due to anisotropies.}

In our previous work,$^{22}$ we have derived an explicit expression for $\Omega$ of a homogeneous system of bosons, described by the Hamiltonian in Eq. (2). Minimization of thermodynamic potential with respect to the phase $\xi$ and condensate fraction $\rho_0$, together with the requirement of dynamical stability of BEC led to the following conclusions (see Table 1 of Ref. 22).

(a) The condensate has a definite phase,$^{23}$ which is independent of temperature or magnetic field.

(b) The phase angle $\Theta$ may have only discrete values, namely $\Theta = \pi n$ and $\Theta = \pi/2 + 2\pi n$ ($n = 0, \pm 1, \pm 2, \ldots$) for an equilibrium system of bosons without and with DM interaction, respectively.

$$2150223-3$$
(c) The presence of a weak DM interaction even with a tiny strength smears out the phase transition from BEC to normal phase into a crossover, i.e., the condensate fraction may vanish only asymptotically by increasing the temperature. Besides, the DM interaction fixes the direction of staggered magnetization, predicted by Matsumoto et al., based on symmetry considerations.

In the present work, we shall study the modification of some physical observables due to EA and DM anisotropies given by Eq. (2).

The rest of this paper is organized as follows. In Sec. 2, we discuss the properties of main equations of the present approach. In Sec. 3, we analyze the role of anisotropies for the thermodynamic parameters such as anomalous density, self-energies, magnetization and heat capacity. We compare our theoretical results with experimental ones for the TlCuCl$_3$ compound in Sec. 4 and summarize our main results in Sec. 5.

Throughout the paper we adopt the units $k_B \equiv 1$ for the Boltzmann constant, $\hbar \equiv 1$ for the Planck constant and $V \equiv 1$ for the unit cell volume. In these units, the energies are measured in Kelvin (K), the mass $m$ is expressed in K$^{-1}$, the magnetic susceptibility $\chi$ for the magnetic fields measured in Tesla (T) has the units of K/T$^2$, while the momentum and specific heat $C_H$ are dimensionless. Particularly, the Bohr magneton is $\mu_B = \hbar e / 2m_0 c = 0.671668$ K/T, where $m_0$ is the free electron mass and $e$ is the fundamental charge.

2. Properties of Main Equations for Self-Energies

One of the main quantities to describe the low-temperature properties of ultracold bosonic systems is the dispersion relation for quasiparticles, which is supposed to be written as $E_k = \sqrt{\varepsilon_k + X_1 \sqrt{\varepsilon_k} + X_2}$, in general. Here, $\varepsilon_k$ is the bare dispersion of triplons and the quantities $X_{1,2}$ are related to the ordinary normal $\Sigma_n$, and anomalous $\Sigma_{an}$, self-energies as follows $X_{1,2} = \Sigma_n \pm \Sigma_{an} - \mu$. The self-energies $X_{1,2}$ and the condensate fraction are the solutions to the following equations:

\begin{align*}
X_1 &= 2U \rho + U\sigma - \mu + \frac{U \rho_0 (\xi^2 + \bar{\xi}^2)}{2} + \gamma + \frac{2\gamma'^2 D_1}{X_2^2}, \\
X_2 &= 2U \rho - U\sigma - \mu - \frac{U \rho_0 (\xi^2 + \bar{\xi}^2)}{2} - \gamma - \frac{2\gamma'^2 D_2}{X_2^2}, \\
\frac{\partial \Omega}{\partial \rho_0} &= \cos 2\Theta (U\sigma + \gamma) + U(\rho_0 + 2\rho_1) - \mu - \frac{\gamma' \sin \Theta \sqrt{\rho_0}}{\sqrt{\rho_0}} = 0,
\end{align*}

where

\begin{align*}
A'_1 &= \frac{\partial A}{\partial X_1} = \frac{1}{8} \sum_k \left( \frac{E_k W'_k + 4W_k}{E_k} \right), \\
A'_2 &= \frac{\partial A}{\partial X_2} = \frac{1}{8} \sum_k \left( \frac{(\varepsilon_k + X_1)^2 (E_k W'_k - 4W_k)}{E_k} \right),
\end{align*}
Effects of exchange and weak DM anisotropies

\[ B'_1 = \frac{\partial B}{\partial X_1} = \frac{1}{8} \sum_k \left( (\varepsilon_k + X_2)^2 (E_k W'_k - 4W_k) \right), \quad (4c) \]

\[ D_1 = \frac{A'_1}{D}, \quad D_2 = \frac{B'_1}{D}, \quad D = A_1'^2 - A_2'B'_1, \quad (4d) \]

\[ W_k = \frac{\coth(\beta E_k/2)}{2}, \quad W'_k = \beta \left( 1 - 4W_k^2 \right) = \frac{-\beta}{\sinh^2(\beta E_k/2)}. \quad (4e) \]

In the above equations, \( A = \rho_1 - \sigma, \) \( B = \rho_1 + \sigma \) and the normal \( \rho_1 \) and anomalous \( \sigma \) densities are given below. In the Hartree–Fock–Bogoliubov approximation, these self-energies play an essential role. Thus, we first study their properties and then evaluate physical observables under consideration. For simplicity, we rewrite Eqs. (3) and (4) separately for the cases with \( (\gamma' = 0) \) and without \( (\gamma' \neq 0) \) DM interactions, taking into account that for these cases \( \xi = \pm 1 \) and \( \xi = i \), respectively.

2.1. Mode 1: \( \gamma' = 0, \gamma \neq 0, \xi = 1 \)

This mode corresponds to the case when only EA is present. Here, we have both phases, BEC and normal, which are sharply separated by the critical temperature \( T_c \) defined by the equation \( \rho_0(T = T_c) = 0 \). The condensate fraction is given in BEC phase by

\[ \rho_0 = \frac{\Delta - 2\gamma - U\sigma}{U}, \quad (5) \]

where the normal \( \rho_1 \) and anomalous \( \sigma \) densities are given by following general expressions

\[ \rho_1 = \sum_k \left[ \frac{W_k (\varepsilon_k + X_1/2 + X_2/2)}{E_k} - \frac{1}{2} \right] \equiv \sum_k \rho_{1k}, \quad (6a) \]

\[ \sigma = \frac{(X_2 - X_1)}{2} \sum_k \frac{W_k}{E_k} \equiv \sum_k \sigma_k \quad (6b) \]

with \( X_1 = 2\Delta, \) \( X_2 = 2\gamma, \) \( E_k = \sqrt{(\varepsilon_k + 2\Delta)(\varepsilon_k + 2\gamma)} \), \( \rho = \rho_0 + \rho_1 \).

In the normal phase \( \rho_0(T > T_c) = 0 \), the self-energies \( X_1 \) and \( X_2 \) in the dispersion relation \( E_k \equiv \omega_k = \sqrt{(\varepsilon_k + X_1)(\varepsilon_k + X_2)} \) are given as follows

\[ X_{1,2}(T > T_c) = 2U\rho - \mu \pm (U\sigma + \gamma), \quad (7) \]

where the total triplon density is

\[ \rho(T > T_c) = \sum_k \frac{1}{e^{\beta \omega_k} - 1}. \quad (8) \]

Explicit expressions for other quantities are moved to Appendix A for convenience.

Note that our mean-field-based main equations are rather general leading to well-known approximations used in the literature for the isotropic case, when all anisotropies are neglected. Particularly, one may derive HFP approximation and simple Bogoliubov approximations as follows.
A. Rakhimov, A. Khudoyberdiev & B. Tanatar

- **HFP approximation.** This is widely used in literature and obtained simply by neglecting $\sigma$ and $\gamma$ in Eq. (5) resulting in the following equation for the condensate fraction

$$\rho_0 = \rho - \rho_1 = \rho - \sum_k \left[ \frac{W_k(\varepsilon_k + U\rho_0)}{\sqrt{\varepsilon_k} \sqrt{\varepsilon_k + 2U\rho}} \right] \frac{1}{2}. \quad (9)$$

- **Bogoliubov approximation.** Further, at zero-temperature, making formal replacement $\rho_0 \rightarrow \rho$ on the right-hand side of Eq. (9) gives

$$\rho_0 = 1 - \frac{1}{2\rho} \sum_k \left[ \frac{\varepsilon_k + U\rho}{\sqrt{\varepsilon_k} \sqrt{\varepsilon_k + 2U\rho}} - 1 \right]. \quad (10)$$

For infinite uniform system with $\varepsilon = \frac{\vec{k}^2}{2m}$, $|k| = 0, \ldots, \infty$, one may evaluate the momentum integration in Eq. (10) to obtain the following well-known formula

$$\rho_0 = 1 - \frac{8\sqrt{\rho a_s^2}}{3\sqrt{\pi}}, \quad (11)$$

where $a_s = Um/4\pi$ is the s-wave scattering length. Remarkably, the quantum depletion given by the second term on the right-hand side of Eq. (11), as well as the energy dispersion in Eq. (10) $E_k = \sqrt{\varepsilon_k \varepsilon_k + 2U\rho}$ were proposed by Bogoliubov more than 70 years ago and has been one of the cornerstones of our understanding of interacting quantum fluids.

2.2. **Mode 2: $\gamma' \neq 0, \gamma \neq 0, \xi = i$**

Here, both EA and DM interactions are present. The main equations for self-energies $X_1$ and $X_2$ are obtained from Eq. (20) of Ref. 22 by setting $\xi = i$,

$$X_1 = 2U\sigma + 2\gamma + \frac{\gamma'}{\sqrt{\rho_0}} + \frac{2\gamma'^2 D_1}{X_2}, \quad (12a)$$

$$X_2 = 2U\rho_0 + \frac{\gamma'}{\sqrt{\rho_0}} - \frac{2\gamma'^2 D_2}{X_2}. \quad (12b)$$

The equation for the condensate fraction $\rho_0$ may be presented in the following dimensionless compact form

$$r_0^3 + Pr_0 + Q = 0, \quad (13)$$

where we have introduced $P = -\bar{\sigma} + 2(\bar{\rho}_1 - 1 - \bar{\gamma})$, $Q = -2\bar{\gamma}'/\sqrt{\bar{\rho}_0}$, $r_0^3 = \rho_0/\rho_c$, $\bar{\sigma} = \sigma/\rho_c$, $\bar{\rho}_1 = \rho_1/\rho_c$, $\bar{\gamma} = \gamma/\mu$, $\bar{\gamma}' = \gamma'/\mu$ in which $\rho_c$ is the critical density of pure BEC, $\rho_c = \mu/2U$.

In general, one has to solve these three coupled nonlinear algebraic equations for the unknown quantities $X_1$, $X_2$ and $r_0$ at a given temperature and magnetic field. Clearly, in such cases it is important to guess the initial values of $X_1(T)$ and $X_2(T)$, since the solutions are not unique. For this purpose it will be convenient to
Effects of exchange and weak DM anisotropies

start from a higher-temperature, say \( T \approx 15 \text{ K} \), where \( \sigma(T \gg T_c) \approx 0, \gamma^2/X_2^2 \rightarrow 0 \) and hence Eqs. (12a) and (12b) are simplified to

\[
Z_1 = \frac{\gamma}{\mu} - \frac{Q}{4r_0}, \quad Z_2 = \frac{r_0^2}{2} - \frac{Q}{4r_0},
\]

where \( Z_1 = X_1/2\mu \) and \( Z_2 = X_2/2\mu \).

2.2.1. High temperatures

For a weak EA interaction, \( \gamma/\mu \ll 1 \) Eq. (14) coincide with those obtained by Sirker et al.\(^\text{28}\) within the HFP approximation with \( \sigma = \gamma = 0 \), and may be solved easily by inserting \( Z_1, Z_2 \) into Eq. (13), thus by reducing the system of three coupled equations into one cubic algebraic equation with respect to \( r_0 \). It is clear that in this regime Eqs. (3a) and (3b) are simplified as follows

\[
X_1(T \gg T_c) \approx X_2(T \gg T_c) = 2U\rho - \mu,
\]

where \( \rho = \rho_0 + \rho_1 \) is the total density of triplons, and \( T_c \) is defined as \( (d\rho/dT)|_{T=T_c}=0 \), \( (d^2\rho/dT^2)|_{T=T_c} \geq 0 \) and hence the normal \( \Sigma_n \) and anomalous \( \Sigma_{an} \) self-energies have the form

\[
\Sigma_n = \mu + \frac{X_1 + X_2}{2} \approx 2U\rho, \quad \Sigma_{an} = \frac{X_1 - X_2}{2} \approx 0.
\]

In Fig. 1, we present typical solutions of Eqs. (12) and (13) as a function of temperature for \( \gamma'=0 \). It is seen that at high temperatures \( X_1 \) and \( X_2 \) overlap with that of pure BEC with \( \gamma = \gamma' = 0 \) in accordance with Eq. (15). Therefore, the effect of anisotropy on self-energies is negligibly small at high temperatures. On the other hand, the effect of DM interaction on the condensate fraction is rather significant, as it is seen from Fig. 1(c).

In fact, since in the presence of DM interaction the parameter \( Q \) is finite, Eq. (13) does not have a zero solution, as illustrated in Fig. 1(c). Strictly speaking, at any temperature there exists a finite condensate fraction. Thus, comparing \( \rho_0(T) \) for pure BEC (dotted curve) with that for the case of DM interaction (solid curve) in Fig. 1(c) one may conclude that DM anisotropy smears out BEC transition into a crossover.

2.2.2. Low temperatures

Moreover, comparing those curves in Fig. 1(c) at low temperatures one may note that the DM interaction enhances the condensate fraction significantly. For example, the condensate fraction at \( T = 0 \) for \( \gamma' = 0.1 \text{ K} \) is nearly 2.7 times larger than that for \( \gamma' = 0 \), corresponding to the isotropic case.

We now discuss the low-temperature behavior of self-energies \( X_1 \) and \( X_2 \). As it is seen from Fig. 1 in this region in the approach by Sirker et al.\(^\text{28}\), \( X_1 \) and \( X_2 \) are nearly of the same order, while in the present approximation \( X_1 \) is much smaller.
Fig. 1. (Color online) Physical solutions of Eqs. (12) and (13) with only anisotropic DM interaction for the input parameters $g = 2.06$, $U = 315$ K, $H = 8.5$ T, $\gamma' = 0.1$ K, and $\gamma = 0$ as a function of temperature. The parameters of bare dispersion $\varepsilon_k$ are taken from Ref. 9. (a)–(c) represent the self-energies $X_1(T)$, $X_2(T)$ and the condensate fraction $\rho_0(T)/\rho_0c$ ($\rho_0c = \mu/2U = 0.07$), respectively, while (d) illustrates the ratio $X_1(T)/X_2(T)$. Solid and dashed lines correspond to present approximation and that by Ref. 28, corresponding to the case with formally setting $\sigma = \gamma = (\gamma')^2 = 0$ in Eqs. (12) and (13), respectively. The dotted lines represent isotropic case with $\gamma = \gamma' = 0$.

than $X_2$ ($X_1/X_2 \approx 10^{-4}$). The main reason of this difference is that in the present approximation the anomalous density has not been neglected, and besides, the DM interaction is taken into account up to the second-order in the strength. Now coming back to the main equations for $X_1$ and $X_2$ one may note that, at low temperatures, $D_1$ in Eq. (12a) given by Eq. (4) becomes negligibly small, while $D_2$ in Eq. (12b) remains finite. Thus, Eq. (12a) with $\gamma = 0$ and the difference $X_2 - X_1$ can be written as follows

$$X_1(T \to 0) \approx 2U\sigma + \frac{\gamma'}{\sqrt{\rho_0}},$$

(17a)

$$X_2 - X_1|_{T \to 0} \approx 2U(\rho_0 - \sigma) - \frac{2\gamma'^2D_2}{X_2^2}.$$  

(17b)

From Eq. (17a) it can be immediately seen that, since $\sigma > 0$, $X_1(T \to 0) \neq 0$ when $\gamma' \neq 0$, that is the gap in the quasi-particle dispersion $E_k = \sqrt{(\varepsilon_k + X_1)(\varepsilon_k + X_2)}$

\footnote{See the next section.}
Effects of exchange and weak DM anisotropies

can never be closed for $\gamma' \neq 0$ (we shall come back to this point in Sec. 4). As to
the difference $X_2 - X_1$, it becomes large i.e., $X_2 \gg X_1$ due to the presence of the
last term in Eq. (17b) with $D_2 > 0$, since lowering the temperature leads also to a
decrease in $X_2$.

2.3. Upper boundary for strength of DM interaction

In our previous work, requiring positiveness of self-energies, $X_1$ and $X_2$, we have
found a boundary condition for the strength of EA interaction as $\gamma \leq U/|\sigma|$. Now
we address the question whether a similar condition be found for the strength of
DM interaction $\gamma'$.

First, we note that Eq. (13) for $r_0$ has a positive solution regardless of the
sign (and value) of the parameter $P$. In fact, since $\gamma' > 0$, the number of sign
changes in this equation is equal to unity, it has exactly one positive solution due
to Descartes’ Rule of Signs. Hence, in the approximation suggested by Ref. 17, the
right-hand side of Eq. (14) is positive for any $\gamma' > 0$. Thus, when we neglect $\sigma$
and use the approximation linear in $\gamma'$, there is no upper bound for the strength of
DM interaction $\gamma'$. However, when we go beyond such an approximation, we have
to deal with Eqs. (3a) and (3b), where the last terms with $\gamma'^2$ play an important
role for large $\gamma'$. By examining the coefficients of $\gamma'^2$, namely $D_1$ and $D_2$ given in
Eq. (4) one may find that $D_2 > 0$ and $D_1 < 0$ at any temperature. Now, it can be
understood that at large values of $\gamma'$, the last term in Eq. (12b) will dominate over
the first and second terms, making the right-hand side of this equation negative.
Actual numerical analysis for TICuCl3 show that this happens at $\gamma' > 0.7K$ for
$H \leq 20$ T. In reality, $\gamma'$ is rather small: $\gamma'_\text{optimum} \approx 0.02$ K (see Sec. 4). Anyway, in
contrast to approximation used in Ref. 17, the present approach, taking into account
$\gamma'$ up to the second-order, is able to predict an upper bound for the strength of DM
interaction $\gamma'_\text{max} \approx 0.7$ K, beyond which this interaction destroys the condensate of
triplons.

3. Sensitivity of Thermodynamic Characteristics to Anisotropies

In Ref. 22, we have shown that the presence of $H_{EA}$ and $D_{DM}$ terms in the bosonic
Hamiltonian with contact interaction may significantly modify the phase and the
condensate fraction of BEC. Now we discuss their influence on some physical
quantities.

3.1. Anomalous density and self-energy

First we show that even a tiny DM interaction changes the sign of anomalous
density $\sigma$, which is negative for pure BEC in finite systems. In fact, subtracting
Eq. (12b) from Eq. (12a) and using Eq. (6b) with $\gamma = 0$, one obtains

$$\sigma = \tilde{S}(\rho_0 - \sigma) - \frac{\tilde{S}_\gamma^2(D_1 + D_2)}{UX_2^2},$$ (18)
Fig. 2. (Color online) (a) The ratio of anomalous density $\sigma$ to the total density $\rho$ of triplons as a function of temperature for various values of DM and EA interactions. (b) The same as in (a) but for the ratio of anomalous and normal self-energies. It is seen that the presence of DM interaction reverses the sign of both anomalous density and self-energy. The solid curves in both figures correspond to isotropic case with $\gamma = \gamma' = 0$. The input parameters are the same as in Fig. 1.

where $\bar{S} = U \sum_k W_k / E_k$. The formal solution of Eq. (18) is

$$\sigma = \frac{\bar{S}[\rho_0 - \gamma'^2(D_1 + D_2)/UX^2]}{1 + \bar{S}}.$$  \hspace{1cm} (19)

Now, from the explicit expressions for $D_1$, $D_2$ defined in Eq. (4) it can be shown that $(D_1 + D_2) \leq 0$. Thus, from Eq. (19) it is understood that $\sigma(\gamma' \neq 0) \geq 0$ at any temperature for $U > 0$, $\gamma' > 0$. Numerical results presented in Fig. 2(a) confirm this conclusion. As to the magnitude of anomalous density, it is seen that both kind of anisotropies lead to increase of $|\sigma|$, which may reach even 20% of the total density of triplons for the moderate values of $\gamma'$.

In Fig. 2(b), a similar quantity, namely, the ratio of anomalous self-energy to the normal self-energy, $\Sigma_{an}/\Sigma_{n}$ is presented. It is seen that $\Sigma_{an}$ does not vanish even in the normal phase, where it is equal to $\Sigma_{an}(T > T_c) = \gamma$. Moreover, the presence of DM interaction changes the sign of $\Sigma_{an}$.

### 3.2. Shift in the critical temperature

The critical temperature $T_c$ is one of the main characteristics of systems undergoing BEC transition. It is understood that the presence of any kind of interaction (or geometry of a trap) modifies the critical temperature of BEC. Quantitatively this is characterized in literature by the relative shift of critical temperature $\Delta T_c/T_c$ defined as

$$\frac{\Delta T_c}{T_c} \equiv \frac{T_c - T_c^0}{T_c^0},$$  \hspace{1cm} (20)

where $T_c^0$ is the critical temperature of BEC transition without the interaction under consideration.

In general, the problem of accurate estimation of the shift turns out to be highly nontrivial, since close to the phase transition, the physics in the interacting gas is
Fig. 3. (Color online) The shift in critical temperature due to EA (a) and DM (b) interactions (solid curves). Dashed curves are phenomenological fits. The input parameters are the same as in Fig. 1.

Effects of exchange and weak DM anisotropies

governed by strong fluctuations, which make perturbation theory inapplicable.\textsuperscript{29} Nevertheless, one can find in the literature some analytical formulas for $\Delta T_c/T_c^0$ due to interparticle contact interaction,\textsuperscript{30} due to the trap geometry,\textsuperscript{31} or due to disorder.\textsuperscript{32,33} We now consider how the critical temperature of triplon BEC may be affected by anisotropies. To find an answer to this question we have to make numerical analysis, since obtaining analytical estimations turns out to be rather complicated.

In Figs. 3(a) and 3(b), we present the dependence of the shift due to EA and DM interactions, respectively. For weak anisotropies these can be approximated in powers of $\gamma/U$ and $\sqrt{\gamma'/U}$ as $\Delta T_c/T_c^0(\gamma) \approx a_1(\gamma/U) + a_2(\gamma/U)^2$ and $\Delta T_c/T_c^0(\gamma') \approx a'_1\sqrt{\gamma'/U} + a'_2(\gamma'/U)$ for the cases of EA and DM interactions, respectively. Clearly the optimized parameters $a_i$ and $a'_i$ depend also on the external magnetic field $H$. Particularly, for TlCuCl$_3$ with $U = 315$ K at $H = 8.5$ T we obtained $a_1/U = 1.167$ K$^{-1}$, $a_2/U^2 = -1.194$ K$^{-2}$, $a'_1/\sqrt{U} = 1.647$ K$^{-1/2}$ and $a'_2/U = 0.053$ K$^{-1}$, as illustrated in Fig. 3.

First, one may note that in both cases $\Delta T_c \geq 0$, which means that presence of the anisotropies shift the critical temperature of BEC transition (or a crossover in the case of DM anisotropy) toward higher values. Second, it is seen that the influence of anisotropy is not negligibly small at moderate values of the intensities. For instance, DM interaction with $\gamma' \approx 0.1$ K modifies $T_c$ with $\Delta T_c/T_c^0(\gamma' = 0.1$ K) $\sim 50\%$. Third, DM anisotropy modifies the critical temperature more strongly than EA anisotropy. For example, for the equal values of intensities, say, $\gamma = \gamma' \approx 0.1$ K, the shift due to DM interaction is nearly five times larger than due to EA interaction. Thus, the critical temperature is more sensitive to DM interaction than to EA.

3.3. Magnetization

In Fig. 4, the uniform magnetization $M(T)$ and $M^2(T)$ are presented for various values of $\gamma$ and $\gamma'$ as a function of temperature. It is seen that the EA interaction...
Fig. 4. (Color online) Uniform magnetization as a function of temperature with only EA (a) and DM (b) anisotropies. Solid lines correspond to the isotropic case with $\gamma = \gamma' = 0$. (c) and (d) display the square of staggered magnetization ($M_\perp^2$). The input parameters are the same as in Fig. 1.

modifies both of these quantities mainly at low temperatures ($T \leq T_c$) [Figs. 4(a) and 4(c)]. As to the DM interaction its effect is twofold. At low temperatures it enhances $M$ as well as $M_\perp$ and in contrast to EA interaction, it prevents the staggered magnetization from vanishing at $T \geq T_c$. Thus, taking into account of DM anisotropy, at least in the linear form as in Eq. (2) within MFA, is inevitable in the accurate description of experimental data on $M_\perp$, reported by Tanaka et al.\textsuperscript{34}

3.4. Heat capacity at constant field $C_H$

In the presence of BEC, the heat capacity exhibits the following specific features.

- Its dependence on temperature has a well-known $\lambda$-shape\textsuperscript{35} which was first observed in superfluid helium.\textsuperscript{36}
- Near absolute zero, $C_V(T)$ behaves like $C_V(T) \propto T^3$, due to a linear energy dispersion, responsible for the superfluidity.

2150223-12
Effects of exchange and weak DM anisotropies

Fig. 5. (Color online) The heat capacity $C_H$ as a function of temperature with only EA anisotropy (a) Solid lines correspond to the isotropic case with $\gamma = \gamma' = 0$. (b) The discontinuity in $C_H$ (upper panel) and $d\rho/dT$ (lower panel) near $T = T_c$. The dashed curve is a phenomenological fit. The input parameters are the same as in Fig. 1. Here, one should note that the presence of anisotropies modifies not only $C_H$ but also $T_c$.

- Near the critical temperature $C_V$ has a discontinuity, i.e., $\Delta C_V \equiv \lim_{\epsilon \to 0} [C_V(T_c - \epsilon) - C_V(T_c + \epsilon)] \neq 0$ which is expected for a second-order phase transition.

In the present work to study these features of the heat capacity of triplons at constant magnetic field and in the presence of anisotropies, we evaluate $C_H(T)$ for the case of only EA anisotropy [see Fig. 5(a)]. First, it is seen that in both cases of low and high temperatures, behavior of $C_H(T)$ is not modified significantly, almost coinciding with the case without anisotropy [solid lines in Fig. 5(a)]. That is the anisotropies are prominent mainly in the critical region. Further, EA interaction leaves the famous $\lambda$-shape almost unchanged [Fig. 5(a)]. Actually, in the presence of EA anisotropy, there is a definite point $T_c$ where $\rho_0(T = T_c) = 0$, which separates BEC and normal phases. This leads to a sharp maximum in the specific heat [see Fig. 5(a)], as in the case of a pure BEC without any anisotropy [solid line in Fig. 5(a)]. In order to find the shift in $\Delta C_H$ due to the EA interaction, we evaluated $\Delta C_H$ as a function of the strength of EA interaction using Eqs. (A.8b), (A.11) and (A.14). The results are presented in Fig. 5(b). It is seen that $\Delta C_H(\gamma = 0, \gamma' = 0) \approx 0.01$ i.e., the discontinuity is positive for a pure BEC, as it is expected. For small values of the EA strength, $0 < \gamma \leq 0.1$ K, the function $\Delta C_H(\gamma)$ can be approximated [dashed line in Fig. 5(b)] by $\Delta C_H(\gamma) \sim b + a\gamma$. For example, at $H = 8.5$ T, the optimal values are $b \approx 0.01$ and $a = 0.055$ K$^{-1}$. In spite of the presence of EA anisotropy, $\Delta C_H$ remains finite which proves that the corresponding BEC-like transition may be classified as a second-order phase transition. From Fig. 5(b) it is seen that $\Delta C_H(\gamma)$ is always positive (upper panel), while $\Delta (d\rho/dT)$ remains negative for any $\gamma$. This is in a good agreement with $C_H$ in the presence of DM anisotropy will be discussed in a separate paper.
4. Results for Realistic Parameters for TlCuCl\textsubscript{3} and Discussions

In the previous section, we studied the effect of anisotropies on thermodynamic quantities. Particularly, we have shown that in contrast to EA interaction, DM interaction modifies their behavior dramatically. It smears BEC transition to a crossover and changes the sign of the anomalous density. Clearly, the significance, or measurability of such effects depend on their interaction strengths $\gamma$ and $\gamma'$. Evidently, unless we have realistic values for these parameters for a real material, our studies will remain purely academic.

Among the 3D quantum dimerized magnets with a spin gap TlCuCl\textsubscript{3} seems to be the most experimentally studied compound.\textsuperscript{2,4,8,9,12,40–50} The observation of a finite $M_\perp$ at $T \geq T_c$\textsuperscript{34} uniquely indicates the presence of DM interaction with a finite $\gamma'$. Thus, using existing experimental data on the magnetization and the heat capacity of TlCuCl\textsubscript{3}, we have made an attempt to obtain optimal values of input parameters of the present approach. The result for $H//b$ is as follows. $g = 2.06$, $U = 367$ K, $\gamma = 0.05$ K and $\gamma' = 0.0201$ K. The magnetizations $M$ and $M_\perp$ corresponding to this set of parameters are depicted in Figs. 6(a) and 6(b), respectively. It is seen that the inclusion of DM anisotropy gives a good description of the staggered magnetization especially at higher temperatures [see, inset of Fig. 6(b)]. Moreover, taking into account the anomalous density $\sigma$ leads to a better description of $M$ e.g., at low temperatures, compared with approximation suggested in Ref. 28, where $\sigma$ has been neglected.
Remarkably, the experimental fit of parameters can be reached with rather small values of anisotropies, namely \(\gamma/U = 1.36 \times 10^{-4}\) and \(\gamma'/U = 5.47 \times 10^{-5}\). In order to compare \(C_H\) with existing experimental data, one needs to perform calculations in the presence of both kinds of anisotropies and solve the problem concerning the extraction of a phonon contribution from experimental curves. This rather complicated task will be the subject of our separated paper.

Thus we have found that the experimental data on magnetization of TlCuCl₃ can be well described by the present approach. On the other hand, there exist experimental measurements on the energy of magnetic excitations. In the following subsection, we shall compare our results with these experiments.

### 4.1. Energy dispersion

As it has been outlined in Sec. 1, a spin-gapped quantum magnet e.g., TlCuCl₃ has a dimer structure and a finite energy gap at zero field \(\Delta_{ST}\) between the singlet \(S = 0\) ground state and the first excited states \(S = 1\). When an external field is applied and reaches a critical value \(H_c = \Delta_{ST}/g\mu_B\) the gap is closed due to the Zeeman effect, as it is illustrated in Fig. 7(a). The excitation spectrum of this compound so far was studied in detail by inelastic neutron scattering (INS)⁴⁴–⁴⁹ as well as ESR measurements.¹²,⁵⁰

The INS studies confirmed that the system becomes quantum critical at \(H_c \approx 5.7\) T where the energy of the lowest Zeeman-split excitation \(|1, -1\rangle\) crosses the nonmagnetic ground state \(|0, +0\rangle\). Above this lowest mode, the system remains in a gapless Goldstone mode and develops a linear dependence on the momentum, which is a good signal of occurrence of BEC. On the other hand, ESR study on this compound gave evidence for a tiny spin gap with minimal value \(\Delta_{an} \approx 0.2\) meV, which was not observed in INS experiments [see Fig. 7(a)]. Therefore, the experimental situation on the energy spectrum of TlCuCl₃ has not been totally clear. In fact, on the one hand, the lowest excitation spectrum for \(H_c \leq H \leq H_{\text{saturation}}\) at \(T \leq T_c\) is gapless, \(\Delta_{an}(\text{INS}) = 0\), on the other hand, it has a finite gap \(\Delta_{an}(\text{ESR}) \neq 0\) and hence cannot be linear. Theoretically, it is clear that if the gap remains finite it may be caused by a lattice anisotropy. Here for clarity, it should be noted that in the present version of mean-field theory one should distinguish two types of energy dispersions. A bare dispersion \(\varepsilon_k \sim k^2/2m\) and the dispersion of collective excitations, given as \(E_k = \sqrt{\varepsilon_k + X_1\sqrt{\varepsilon_k} + X_2}\), where the self-energies \(X_1\) and \(X_2\) are discussed in Sec. 2. The dispersion of elementary excitations at zero field \(\varepsilon_k\) is well studied experimentally⁴⁵,⁴⁷ and presented as a function of momentum and intra (inter)-dimer interactions \(J_i\) as \(\varepsilon_k(J_i)\). One can find in the literature an explicit expression for \(\varepsilon_k(J_i)\) with its optimized parameters,⁴,⁹,⁴⁷ which has also been used in the present work with the normalization \(\varepsilon_k \mid_{k \rightarrow 0} = \vec{k}^2/2m\).³⁹

As to the energy spectrum at \(H \geq H_c\), it is clearly model dependent. For example, in the isotropic case for \(T \leq T_c\) it is gapless, given by \(E_k = \sqrt{\varepsilon_k + X_1\sqrt{\varepsilon_k}} \sim ck + O(k^3)\), thus, \(\Delta_{an} = E_k|_{k \rightarrow 0} = 0\) in agreement with
Fig. 7. (Color online) (a) The schematic illustration of energy levels of a spin-gapped system. At $H = H_c$ the gap $\Delta_{ST}$ closes and may reopen due to anisotropies with a tiny gap $\Delta_{an}$. (b) Energy dispersion of the low-lying magnetic excitations in TlCuCl$_3$. The solid, dashed, and dotted lines correspond to the present approximation including anisotropies; approximation by Ref. [28] and without anisotropy, respectively. The experimental data are taken from Ref. [44].

In the presence of anisotropies it has a finite gap $\Delta_{an} = \sqrt{X_1 X_2}$, where $X_1$ and $X_2$ are defined by Eqs. (12) and (13). Using our optimal input parameters we obtained a finite but rather small value $\Delta_{an}(H = 14 \, T, T = 1.5 \, K) = 10^{-4} \, \text{meV}$, which is consistent with INS measurements, but not with ESR: $\Delta_{an}(H = 14 \, T, T = 1.5 \, K) = 0.2 \, \text{meV}$. In Fig. 7(b), we present quasi-particle spectrum $E_k = \sqrt{\varepsilon_k + X_1 \varepsilon_k + X_2}$ for $H = 14 \, T$ at $T = 1.5 \, K$. It is seen that the excitation energy in the present approximation is almost linear, in accordance with experimental results. However, the experimental values of $E_k^{\text{exp}}$ are rather underestimated. This can be understood as follows. As it has been shown in Sec. 2 at low temperatures, the self-energies especially $X_1$ is rather small (Fig. 1). Our input parameters optimized by experimental magnetizations lead to much smaller values: $X_1(H = 14 \, T, T = 1.5 \, K) = 0.67 \times 10^{-5} \, \text{K}$, $X_2(H = 14 \, T, T = 1.5 \, K) = 0.19 \, \text{K}$, thus $X_1 \ll X_2$. As a result, the momentum dependence of the dispersion is similar to that of isotropic one, $E_k = \sqrt{\varepsilon_k + X_1 \varepsilon_k + X_2} \sim \sqrt{\varepsilon_k + X_2}$ which is practically nothing but the Goldstone mode. Thus, we may come to the conclusion that in accordance with present approximation the lowest excitation energy of TlCuCl$_3$ at very low temperatures has a rather small, but finite gap and exhibits, practically, a linear dispersion at small momentum, in spite of the presence of EA and DM interactions. Note that, a similar situation has been observed for compounds Sr$_3$Cr$_2$O$_8$ and Ba$_3$Cr$_2$O$_8$ which have DM interaction, but no anisotropy gap, i.e., $\Delta_{an}(\text{Sr}_3\text{Cr}_2\text{O}_8) = 0$, $\Delta_{an}(\text{Ba}_3\text{Cr}_2\text{O}_8) = 0$.10

4.2. Discussion

In the present section, having fixed the parameters of the theory by magnetization data on TlCuCl$_3$, we have studied its energy spectrum above the critical field at
Effects of exchange and weak DM anisotropies

$T \leq 1.5$ K. We have found that the description of magnetizations for $H//b$ is quite good, while that of the energy dispersion of the low-lying magnetic excitations needs to be improved. In some sense, this brings to mind the situation in nuclear physics: one can choose optimal parameters for the nucleon-nucleon potential by experimental data on cross sections, but fails to accurately describe the binding energies of light nuclei. Anyway, the main reason of our failure seems to be the simplicity of the Hamiltonian $H_{DM}$ used here (the last term in Eq. (2)). In fact, in deriving this linear Hamiltonian it has been assumed that the DM vector is parallel to $x$, i.e., $\vec{D} = [D_x, 0, 0]$. Therefore, it is naturally expected that by using a more general form for $H_{DM}$, where other components of $\vec{D}$ are also included one will be able to describe not only magnetizations, but also excitation energies in the extended version of the present mean-field approach. Note that by neglecting the other components of the DM vector, one cannot describe magnetizations for $H \perp (1, 0, 2)$ either.

5. Summary and Conclusion

We have studied effects of lattice anisotropies on thermodynamic characteristics of spin-gapped quantum magnets for $H_c \leq H < H_{Saturation}$ by applying our extended mean-field-based approach, proposed in our previous work. This nonperturbative approach takes into account the anomalous density and both EA and DM interactions more accurately than it is done e.g., in the HFP approximation. We derived explicit expressions for some thermodynamic quantities which include the self-energies $X_1$ and $X_2$, and the condensate fraction $\rho_0$. Analysis of the coupled equations with respect to these three quantities show that at high temperatures $T \gg T_c$, the self-energies $X_{1,2}$ are not significantly affected by EA and DM interactions. Meanwhile, the latter strongly modifies the condensate fraction converting BEC transition into a crossover.

At low temperatures the DM interaction increases $\rho_0$, but leads to rather small values of $X_1$, compared with the isotropic case. As a result, the energy dispersion $E_k = \sqrt{(\varepsilon_k + X_1)(\varepsilon_k + X_2)}$, develops a linear dependence at small momentum, in accordance with experimental measurements.

In contrast to EA interaction, the presence of DM interaction, even in the simple linear form in the Hamiltonian, modifies the anomalous density, changing its sign. Particularly, it is expected that the usual “$\lambda$-shape” of the heat capacity disappears due to strong DM interactions. On contrary, the presence of only EA anisotropy leaves the “$\lambda$-shape” of the heat capacity unchanged. The discontinuity in $C_H$ close to the critical temperature is shifted significantly for moderate values of the intensity of the EA.

We have found optimal input parameters of the Hamiltonian for the compound TlCuCl$_3$ which describes experimental data on magnetizations, at least for $H//b$, quite well. This set of parameters lead to a linear dispersion of energy of
A. Rakhimov, A. Khudoyberdiev & B. Tanatar

quasiparticles, but predicts a fairly small value of an anisotropy gap, estimated by ESR measurements.

In future work, we plan to extend our Hamiltonian by taking into account a more realistic DM interaction to obtain better description of experimental data on the spectrum of low lying excitations, as well as the heat capacity.

Acknowledgments

We are indebted to Andreas Schilling for useful discussions and comments. AR acknowledges support by TUBITAK-BIDEB (2221), AK is supported by the Ministry of Innovative Development of the Republic of Uzbekistan and thankful to group of J. Osterwalder at Physics Institute of University of Zurich. BT is supported by Science and Technological Council of Turkey (TUBITAK) under Grant No: 119N689 and Turkish Academy of Sciences (TUBA) under Grant No. AD21. This work is partly supported by funding from Academy of Sciences of the Republic of Uzbekistan.

Appendix A. Explicit Expressions for Some Thermodynamic Parameters

As shown in Sec. 2, the physics of the cases with and without anisotropies are quite different. In the presence of DM interaction, all useful expressions for physical observables may be found by setting $\xi = i$ in Eqs. (3a) and (3b) which are to be solved with the restrictions $X_1 \geq 0$, $X_2 \geq 0$. However, when DM is absent ($\gamma' = 0, \gamma \neq 0$), one must be aware of the Hugenholtz–Pines (HP) theorem\(^\text{51}\) which holds in the limit $\gamma \to 0$. We discuss these two cases separately as follows.

A.1. Mode 1: $\gamma' = 0, \gamma \neq 0$

We start from the explicit expression for $\Omega$,

$$
\Omega(\gamma' = 0, \gamma \neq 0, \xi = 1) = U \rho_1^2 + \frac{U(\sigma^2 + \rho_0^2)}{2} + \rho_1 \left( \frac{X_1}{2} - \frac{X_2}{2} - \mu + 2U\rho_0 \right) + \sigma \left( \frac{X_2}{2} - \frac{X_1}{2} + \gamma + U\rho_0 \right) + \gamma\rho_0 - \mu_0\rho_0 + \Omega_T, \quad (A.1)
$$

where

$$
\Omega_T = \frac{1}{2} \sum_k (E_k - \varepsilon_k) + T \sum_k \ln(1 - e^{-\beta E_k}), \quad (A.2a)
$$

$$
X_1 = U(3\rho_0 + 2\rho_1 + \sigma) - \mu + \gamma, \quad (A.2b)
$$

$$
X_2 = U(\rho_0 + 2\rho_1 - \sigma) - \mu - \gamma, \quad (A.2c)
$$

$$
E_k = \sqrt{(\varepsilon_k + X_1)(\varepsilon_k + X_2)} \quad (A.2d)
$$
Effects of exchange and weak DM anisotropies

and \( \mu_0 = 2U \rho_1 + U \sigma + \gamma + U \rho_0 \) is introduced to avoid the Hohenberg–Martin dilemma\(^{52} \) in the condensate phase. In this phase, \( \rho_0(T \leq T_c) = 0 \) and HP relation may be written in a slightly “broken” form\(^{25} \):

\[
\Sigma_n - \Sigma_an - \mu = X_2 = 2\gamma
\]

(A.3)

which gives a gapless energy dispersion in the \( \gamma \to 0 \) limit:

\[
E_k(T < T_c) = \sqrt{(\epsilon_k + X_1)(\epsilon_k + X_2)}
\]

(A.4)

whose solution is positive definite due to \( |\rho_0| \geq |\sigma| \). This equation may be rewritten in a more convenient form as

\[
\Delta = \frac{X_1}{2} = \mu + 2U(\sigma - \rho_1) + 5\gamma,
\]

(A.5)

where \( \sigma \) and \( \rho_1 \) are given by Eqs. (6a) and (6b) with \( X_2 = 2\gamma \), \( X_1 = 2\Delta \) and \( \mu = g\mu_B(H - H_c) \). Having solved Eq. (A.5) with respect to \( \Delta \), one may evaluate the densities as follows

\[
\rho_0 = \frac{\Delta - 2\gamma - U\sigma}{U},
\]

(A.6a)

\[
\rho = \frac{\Delta + \mu + \gamma}{2U}.
\]

(A.6b)

In the normal phase \( (T > T_c) \), one may neglect \( \rho_0 \) in Eqs. (A.2b) and (A.2c) to obtain

\[
X_{1,2}(T > T_c) = 2U\rho - \mu \pm \gamma \pm \sigma,
\]

(A.7a)

\[
\rho(T > T_c) = \sum_k \frac{1}{e^{\beta \omega_k} - 1},
\]

(A.7b)

\[
\omega_k = \sqrt{(\epsilon_k + X_1)(\epsilon_k + X_2)}.
\]

(A.7c)

The entropy \( S \), heat capacity \( C_H \) and Grüneisen parameter may be found as follows\(^{53,54} \)

\[
S = -\left( \frac{\partial \Omega}{\partial T} \right)_H = -\sum_k \ln[1 - \exp(-\beta \epsilon_k)] + \beta \sum_k \frac{\epsilon_k}{e^{\beta \epsilon_k} - 1},
\]

(A.8a)

\[
C_H = T\left( \frac{\partial S}{\partial T} \right)_H = \frac{1}{4} \sum_k W_k \epsilon_k (\epsilon_k' - \beta \epsilon_k),
\]

(A.8b)

\[
\Gamma_H = -\frac{1}{C_H} \left( \frac{\partial \mu}{\partial T} \right)_H = \frac{g\mu_B}{C_H} \left( \frac{\partial \rho}{\partial T} \right)_H,
\]

(A.8c)

where \( \epsilon_k = \sqrt{(\epsilon_k + X_1)(\epsilon_k + X_2)} \), \( \epsilon_k' = (\partial \epsilon_k/\partial T)_H \) and \( X_{1,2} \) are given by Eqs. (A.4) and (A.7a). We give explicit expressions for \( \epsilon_k' \) and \( \rho'_T \) for normal \( (T > T_c) \) and BEC \( (T \leq T_c) \) phases in the following where the critical temperature is defined at the point \( \rho_0(T = T_c) = 0 \).
A. Rakhimov, A. Khudoyberdiev & B. Tanatar

A.2. Critical temperature and density

The condition \( \rho_0(T = T_c) = 0 \) leads the following coupled equations with respect to \( T_c \) and \( \sigma_c \):

\[
\frac{\mu}{2U} + \frac{\sigma_c + 3\bar{\gamma}}{2} - \sum_k \frac{f_k(E_k^c)}{E_k^c} [\varepsilon_k + U(\sigma_c + 3\bar{\gamma})] = 0,
\]

\[
\sigma_c + U(\sigma_c + \bar{\gamma}) \sum_k \frac{f_k(E_k^c)}{E_k^c} = 0,
\]  \hspace{1cm} (A.9)

where \( E_k^c = E_k(T = T_c) = \sqrt{\varepsilon_k + X_1^c} / (\varepsilon_k + 2\bar{\gamma}) \), \( X_1^c = 2U(\sigma_c + 2\bar{\gamma}) \), \( f_k(x) = 1/(\exp(x/T_c) - 1) \) and \( \bar{\gamma} = \gamma / U \). The critical density is given by

\[
\rho(T = T_c) = \frac{\mu}{2U} + \frac{\sigma_c + 3\bar{\gamma}}{2} \equiv \rho_c.
\]  \hspace{1cm} (A.10)

A.2.1. Normal phase

For \( T > T_c \), differentiating Eq. (A.7b) and using Eq. (A.7c) we obtain following set of equations:

\[
\rho'_T(T > T_c) = \frac{b_1 a_{22} - b_2 a_{12}}{a_1 a_{22} - a_{12} a_{21}}, \quad \sigma'_T(T > T_c) = \frac{b_2 a_{11} - b_1 a_{21}}{a_1 a_{22} - a_{12} a_{21}},
\]

\[
\frac{d\omega}{dT} = \frac{U}{\omega_k} [2(\varepsilon_k - \mu + 2U\rho)\rho'_T - U(\bar{\gamma} + \sigma)\sigma'_T],
\]

\[
a_{11} = 1 - 2A + \frac{U}{2} \sum_k (\varepsilon_k - \mu + 2U\rho)^2 (-2 + 4W_k - \omega_k W'_k),
\]

\[
a_{12} = \frac{U^2}{4} \sum_k (\varepsilon_k - \mu + 2U\rho)(\sigma + \bar{\gamma})(2 - 4W_k + \omega_k W'_k),
\]

\[
a_{22} = 1 - \frac{U^3}{4(1 + A)^2} \sum_k \bar{\gamma}(\sigma + \bar{\gamma})(2 - 4W_k + \omega_k W'_k),
\]

\[
a_{21} = \frac{2\bar{\gamma} a_{12}}{(1 + A)^2}, \quad b_1 = -\frac{1}{4T} \sum_k (\varepsilon_k - \mu + 2U\rho)W'_k,
\]

\[
b_2 = \frac{U\bar{\gamma}}{4T(1 + A)^2} \sum_k W'_k, \quad A = U \sum_k \omega_k (\exp(\beta\omega_k) - 1),
\]

where \( W'_k = -\beta / \sinh^2(\beta\omega_k / 2) \) and \( \omega_k = 1/2 \coth(\beta\omega_k / 2) \).

A.2.2. BEC phase

In this case \( \varepsilon_k(T) = E_k(T) = \sqrt{(\varepsilon_k + 2\Delta(T))(\varepsilon_k + 2\bar{\gamma})} \) differentiation of which gives

\[
\varepsilon'_{k,T} = \frac{E'_k}{E_k} = \frac{\varepsilon_k + 2\bar{\gamma}}{E_k} \Delta'_T.
\]  \hspace{1cm} (A.12)
To find $\Delta'_T = (\partial \Delta/\partial T)$, we can differentiate both sides of Eq. (A.5) with respect to $T$ and solve it for $\Delta'_T$. The result is

$$\Delta'_T = \left( \frac{\partial \Delta}{\partial T} \right)_H = \frac{US_3}{2T(2S_4 + 1)}$$

with $S_3 = \sum_k W_k^4(\varepsilon_k + 2\Delta)$ and $S_4 = U \sum_k (4W_k + E_k W'_k)/4E_k$. As to $\rho'_T$ it can be found directly from (A.6b) as follows

$$\rho'_T(T \leq T_c) = \frac{S_3}{4T(2S_4 + 1)}.$$  

At last, setting in Eqs. (A.11) and (A.14) $\rho_0 = 0$, $T = T_c$, $E_k = \omega_k = \sqrt{\varepsilon_k + X_1^4(\varepsilon_k + X_2^4(T))}$, one may define the cusp in $\rho'_T$ as $\Delta \rho'_T = \rho'_T(T_c) - \rho'_T(T_c^+)$ presented in Fig. 5(b). As to the cusp in $C_H$, presented also in Fig. 5(b) may be found in a similar way from Eqs. (A.8b), (A.11) and (A.14).

A.3. Mode 2: $\gamma' \neq 0, \gamma \neq 0$ case

The expressions for $S, C_H$ and $\Gamma_H$ remain formally unchanged. However, explicit expressions for $E'_k, T$ and $\partial \rho/\partial T$ in Eqs. (A.8b) and (A.8c) are quite complicated. Implicit differentiation of $E_k = \sqrt{(\varepsilon_k + X_1^4(T))(\varepsilon_k + X_2^4(T))}$ gives

$$E'_k = \frac{\partial E_k}{\partial T} = \frac{(\varepsilon_k + X_2^4)X_1^4 + (\varepsilon_k + X_1^4)X_2^4}{2E_k},$$

where $X'_1 = \partial X_1/\partial T$ and $X'_2 = \partial X_2/\partial T$ whose explicit expressions will be given below. Now, differentiating both sides of equations $\rho_1 = (A + B)/2$ and $\sigma = (B - A)/2$ with respect to temperature one obtains

$$\frac{d\rho_0}{dT} = C_\rho[X'_1(B'_1 + 3A'_1) + X'_2(A'_1 + 3A'_2) + A'_1 + B'_1],$$

$$\frac{d\rho_0}{dT} = \frac{X'_1}{2}(A'_1 + B'_1) + \frac{X'_2}{2}(A'_1 + A'_2) + \frac{1}{2}(A'_1 + B'_1),$$

$$\frac{d\sigma}{dT} = \frac{X'_1}{2}(B'_1 - A'_1) + \frac{X'_2}{2}(A'_1 - A'_2) + \frac{1}{2}(B'_1 - A'_1),$$

$$\frac{d\rho}{dT} = \frac{d\rho_0}{dT} + \frac{d\rho_1}{dT},$$

where $C_\rho = -U\rho_0^3/(\gamma' + 2U\rho_0^3/2)$, $A'_1 = -(\beta/4) \sum_k W_k^4(\varepsilon_k + X_1^4)$, $B'_1 = -(\beta/4) \sum_k W_k^4(\varepsilon_k + X_2^4)$ and $A'_1 = \partial A/\partial X_1$, $B'_1 = \partial B/\partial X_1$ given in Eq. (4e).

In the above equations $X'_1 = dX_1/dT$ and $X'_2 = dX_2/dT$ are still unknown. To find them we rewrite Eqs. (29a) and (29b) in our previous paper\textsuperscript{22} in the following equivalent form

$$M_{11}A'_1 + M_{12}B'_1 = 0,$$

$$M_{12}A'_1 + M_{11}A'_2 + \frac{2\gamma'}{\sqrt{\rho_0}} = 0,$$

with $M_{11} = -X_2 + 2U\rho_0 + \gamma'/\sqrt{\rho_0}$, $M_{12} = -X_1 + 2U\sigma + 2\gamma' + \gamma'/\sqrt{\rho_0}$.

2150223-21
A. Rakhimov, A. Khudoyberdiev & B. Tanatar

Now, differentiating both sides of Eqs. (A.17) and (A.18) and solving the resulting equations for $X'_1$ and $X'_2$ one finally gets

$$X'_1 = \frac{A_{12}b_2 - b_1A_{22}}{A_{11}A_{22} - A_{21}A_{12}}, \quad X'_2 = \frac{A_{21}b_1 - b_2A_{11}}{A_{11}A_{22} - A_{21}A_{12}},$$

(A.19)

where

$$\begin{align*}
A_{11} &= A'_1 M_{11,1} + M_{11}A'_1 + B'_1 (-1 + M'_{12,2}) + M_{12}B'_1, \\
A_{12} &= A'_1 (-1 + M'_{11,1}) + M_{11}A'_2 + M_{12}B'_1, \\
A_{21} &= A'_1 (-1 + M'_{12,2}) + M_{12}A'_1 + M'_{11,1}A'_2 + M_{11}A'_2, \\
A_{22} &= A'_1 M_{12,2} + M_{12}A'_2 + (-1 + M'_{11,1})A'_2 + M_{11}A'_2, \\
b_1 &= M'_{11,1}A'_1 + M_{11}A'_{1,t} + M_{12}B'_1 + M_{12}B'_{1,t}, \\
b_2 &= M'_{12,2}A'_1 + M_{12}A'_{2,t} + M'_{11,1}A'_{2} + M_{11}A'_{2,t},
\end{align*}$$

(A.20a-d)

and we have introduced the abbreviations $M'_{i,j,k} = \partial M_{ij}/\partial X_k$ and $A''_{i} = \partial^2 A/\partial X_1 \partial X_j$. $f'_t$ is an explicit derivative with respect to temperature $f'_t(\varphi(X_1(T), X_2(T), T) = df/dT - (\partial f/\partial \varphi)X'_1 - (\partial f/\partial \varphi)X'_2$.

References

1. V. Zapf, M. Jaime and C. D. Batista, Rev. Mod. Phys. 86, 563 (2014).
2. E. Ya. Sherman et al., Phys. Rev. Lett. 91, 057201 (2003).
3. X.-G. Zhou et al., Phys. Rev. Lett. 125, 267207 (2020).
4. M. Matsumoto et al., Phys. Rev. Lett. 89, 077203 (2002).
5. V. I. Yukalov, Laser Phys. 22, 1145 (2012).
6. J. O. Andersen, Rev. Mod. Phys. 76, 599 (2004).
7. N. Cavadini et al., Eur. Phys. J. B 7, 519 (1999).
8. M. Matsumoto et al., Phys. Rev. B 69, 054423 (2004).
9. G. Misguich and M. Oshikawa, J. Phys. Soc. Jpn. 73, 3429 (2004).
10. Z. Wang et al., Phys. Rev. B 89, 174406 (2014).
11. A. K. Kolezhuk et al., Phys. Rev. B 70, 020403(R) (2004).
12. V. N. Glazkov et al., Phys. Rev. B 69, 184410 (2004).
13. V. N. Glazkov et al., Phys. Rev. B 85, 054415 (2012).
14. K. Yu. Povarov et al., Phys. Rev. Lett. 107, 037204 (2011).
15. S. A. Zvyagin et al., Phys. Rev. Lett. 98, 047205 (2007).
16. S. Miyahara et al., J. Phys.: Condens. Matter 16, 911 (2004).
17. J. Sirker, A. Weisse and O. P. Sushkov, Europhys. Lett. 68, 275 (2004).
18. A. Rakhimov, S. Mardonov and E. Ya. Sherman, Ann. Phys. 326, 2499 (2011).
19. F. Cooper et al., Phys. Rev. A 83, 053622 (2011).
20. H. Kleinert and V. Schulte-Frohlinde, Critical Properties of $\phi^4$-Theories (World Scientific, Singapore, 2001).
21. A. Rakhimov et al., Phys. Rev. A 77, 033626 (2008).
22. A. Rakhimov et al., Ann. Phys. 424, 168361 (2021).
23. S. M. Barnett, K. Burnett and J. A. Vaccaro, J. Res. Natl. Inst. Stand. Technol. 101(4), 593 (1996).
24. M. Matsumoto, T. Shoji and M. Koga, J. Phys. Soc. Jpn. 77, 074712 (2008).
Effects of exchange and weak DM anisotropies

25. A. Khudoyberdiev, A. Rakhimov and A. Schilling, *New J. Phys.* **19**, 113002 (2017).
26. N. N. Bogoliubov, *J. Phys.* **11**(1), 23 (1947).
27. R. Lopes *et al.*, *Phys. Rev. Lett.* **119**, 190404 (2017).
28. J. Sirker, A. Weiss and O. P. Sushkov, *J. Phys. Soc. Jpn.* **74**, 129 (2005).
29. V. I. Yukalov and E. P. Yukalova, *Laser Phys. Lett.* **14**, 073001 (2017).
30. F. F. de Souza Cruz, M. B. Pinto and R. O. Ramos, *Phys. Rev. B* **64**, 014515 (2001).
31. P. Arnold and B. Tomasik, *Phys. Rev. A* **64**, 053609 (2001).
32. A. Rakhimov *et al.*, *New J. Phys.* **14**, 113010 (2012).
33. A. V. Lopatin and V. M. Vinokur, *Phys. Rev. Lett.* **88**, 235503 (2002).
34. H. Tanaka *et al.*, *J. Phys. Soc. Jpn.* **70**, 939 (2001).
35. K. Huang, *Statistical Mechanics* (John Wiley & Sons, 1987).
36. R. W. Hill and O. V. Lounasmaa, *The Specific Heat of Liquid Helium* (Philosophical Magazine, 1957).
37. L. D. Landau and E. M. Lifshitz, *Statistical Physics*, 3rd edn., Part 1 (Elsevier Butterworth-Heinemann, 1980).
38. A. Rakhimov and I. N. Askerzade, *Int. J. Mod. Phys. B* **29**(18), 1550123 (2015).
39. A. Rakhimov *et al.*, *Phys. Rev. B* **98**, 144416 (2018).
40. R. Dell’Amore, A. Schilling and K. Krämer, *Phys. Rev. B* **78**, 224403 (2008).
41. A. Furrera and C. Rüegg, *Physica B* **385**, 295 (2006).
42. A. Oosawa, H. Aruga Katori and H. Tanaka, *Phys. Rev. B* **63**, 134416 (2001).
43. A. Oosawa, M. Ishii and H. Tanaka, *J. Phys.: Condens. Matter* **11**, 265 (1999).
44. Ch. Rüegg *et al.*, *Nature* **423**, 62 (2003).
45. A. Oosawa *et al.*, *Phys. Rev. B* **65**, 094426 (2002).
46. Ch. Rüegg *et al.*, *Phys. Rev. Lett.* **95**, 267201 (2005).
47. N. Cavadini *et al.*, *Phys. Rev. B* **63**, 172414 (2001).
48. N. Cavadini *et al.*, *Phys. Rev. B* **65**, 132415 (2002).
49. Ch. Rüegg *et al.*, *Appl. Phys. A* **74**(Suppl.), S840 (2002).
50. S. Kimuraa *et al.*, *J. Magn. Magn. Mater.* **310**, 1218 (2007).
51. N. M. Hugenholtz and D. Pines, *Phys. Rev.* **116**, 489 (1959).
52. P. C. Hohenberg and P. C. Martin, *Ann. Phys.* **34**, 291 (1965).
53. A. Rakhimov, M. Nishanov and B. Tanatar, *Phys. Lett. A* **384**, 126313 (2020).
54. A. Rakhimov *et al.*, *Int. J. Mod. Phys. B* **35**, 2150018 (2021).