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LOW TEMPERATURE PHASE TRANSITIONS IN ANISOTROPIC QUANTUM MAGNETS

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Contents

INTRODUCTION (thesis annotation)

Topicality and relevance of the theme of the dissertation. Nowadays, great attention is paid to solving the important problems of magnetic materials through the study of their phase transitions at low temperatures. Understanding of physics of quantum magnets is very promising, but also quite challenging. It is essential to apply Bose particle physics for quantum magnets and to study phase transitions by quantum effects. Although the Bose–Einstein condensate (BEC) has been observed with bosonic atoms in liquid helium and cold gases, the concept is much more general. Low temperature properties of some quantum magnets can be explained by quantum effects such as by BEC theory. In particular, in antiferromagnets, elementary excitations are magnons, quasiparticles with integer spin and obey Bose statistics. However, the analogy between spins and bosons has proven to be very fruitful in those antiferromagnets where closely spaced pairs of spins form dimers with a spin-singlet ground state and triplet bosonic excitations called triplons. Triplons have a high density and small mass compared to atomic gases. As a result, in this system, condensate can be present at high temperatures in contrast to nano kelvins in atomic gases.

At present, it is also interesting to know the low temperature properties of spin-gapped magnets with long range interactions, including for instance exotic phases or phase transitions and anisotropy. Its results could be used for creating quantum computers and quantum information storage. It has been recently discovered that low temperature magnetic memory of antiferromagnetics is at least 100 times denser than today's hard disk drives and solid-state memory chips. So, investigation of quantum magnets may predict their new properties, which make easier and optimal their application in computer and information technology.

In recent years, great attention has been paid in our country to conducting theoretical and experimental research on magnetic materials and their properties at the world level. Therefore, the study of Bose-Einstein condensate and related problems of magnetic quasiparticles in quantum antiferromagnets at low temperatures is a topical issue. It is important to conduct scientific research at solving fundamental and practical issues in accordance with the tasks set by the Address of the President of the Republic of Uzbekistan to the Oliy Majlis[1] on December 29, 2020.

In recent years, Uzbekistan has been paying great attention to conducting world-class theoretical and experimental research on magnetic materials and their properties. Theoretical and practical research is being conducted to create a permanent Teslali permanent magnetic field and to study the optical properties and nanostructure of samples using this magnetic field. These fundamental researches are of great importance in the development of science in our country.

This dissertation research is based on the Decree of the President of the Republic of Uzbekistan No. PF-4947 of February 7, 2017 "On the Action Strategy for further development of the Republic of Uzbekistan", No. PP-2789 of February 17, 2017. and on measures to finance ", PQ-4526 dated November 21, 2019" On measures to support research activities of the Institute of Nuclear Physics ", as well as to a certain extent in the implementation of the tasks set out in other regulations related to this activity serves.

Relevance of the research to the priority areas of science and technology development of the Republic. The dissertation research was carried out in accordance with the priority areas of science and technology development of the Republic: II. "Power, energy and resource saving".

Degree of study of the problem. Currently, numerous theoretical and practical research on low temperature physics and quantum magnets is being conducted by the world's leading scientists. In particular, Swiss scientists A. Schilling and R. Dell'Amore studied low-phase phase transitions and magnetization properties in quantum magnets using experimental experiments.

[1] Mirziyoev Sh.M. Address of the President of the Republic of Uzbekistan to the Oliy Majlis // People's Speech, December 30, 2020, No. 275-276 (7746-7747),2-p.

Considering the anisotropic properties of quantum magnets, quantum phase transitions and the condensate state of magnons were theoretically studied by Australian scientists (J. Sirker, A. Weisse, O.P. Sushkov) using the Hartree-Fock-Popov approximation.

Italian (V. Zapf, M. Jaime, CD Batista), Russian (VI Yukalov), Japanese (F. Yamada, T. Ono, H. Tanaka, G. Misguich, M. Oshikawa, T. Sakakibara), Uzbek (A. Rakhimov, B. Boyzakov, F. Abdullayev, S. Djumanov, U. Valiev, R. Galimzyanov, E. Quvondiqov) and other researchers conducted many theoretical and experimental studies on condensed matter physics and the study of quantum magnets.

More than a hundred years ago Kamerlingh Onnes experimentally observed that liquid ^{4}He, when cooled below 2.2 K temperature, began to expand unusually. In 1938 Kapitza recognized this effect as the onset of superfluidity, which was later related to the fundamental phenomenon of Bose-Einstein condensation. Since the experimental realization of BEC in dilute trapped atomic gases in 1995 by the JILA group, the BEC became one of the main research fields in modern physics and stimulated research of BEC of quasiparticles such as excitons and polaritons in semiconductors, and triplons in quantum magnets. This great attention is mainly due to beautiful experiments with trapped atoms, optical lattices, optically excited semiconductors, and quantum magnets, accomplished in many laboratories of different countries and promising a variety of interesting applications in nanophysics. A new stage in this field was sparked in 1998 when Massachusetts Institute of Technology (MIT) group realized an optically trapped BEC of the spin -1 ^{23}Na. In contrast to a magnetic trap, in which spin degrees of freedom are frozen, an optical trap can confine atoms in all magnetic sublevels of spin, allowing to study of properties of BECs. Therefore, ultra cold atomic physics has supplied us with a new family of quantum fluids, so-called spinor Bose gases which are subject to the interplay of magnetism and superfluidity. In 1999 A. Oosawa studied magnetizations in magnetic materials to find the critical temperature of the magnetic ordering in quantum antiferromagnetic sample $TlCuCl_3$ below its Neel temperature. The interpretation of experiments calls the development of fundamental theory. Only a correct theory allows for the proper understanding of experiments, can suggest appropriate and realistic technical applications, and predict new properties and phases of matter. It is interesting to note that in condensed matter physics, where collective phenomena such as the BEC involve thousands of particles, experiments advance the theory. This is quite different to the high energy particle physics.

There is another problem in the description of properties of pure quantum magnets in terms of triplon BEC. This problem is related to the geometry of the unit cell. In fact, the interaction between spin dimers hosting triplon states causes anisotropy, i.e. breaking of rotational symmetry of the bosonic system. As a result, the Hamiltonian does not commute with the corresponding particle number operator, making it not a well-defined quantity. This fact causes natural doubts in theoretical description in terms of the BEC. However, it has been shown that, when the anisotropy is relatively small, mean-field approximation (MFA) with a BEC scenario can be used by taking into account the resulting U(1) symmetry breaking term perturbatively. Additionally, there are some open questions which needs to be studied briefly. For example: can we find a description of the effect of anisotropy on the magnetization in the 'noncondensed and condensed phase' (at present only poorly understood), which is at the same time compatible with the theory for the BEC phase with disorder?

Connection of the topic of dissertation with the scientific researches of the higher educational institutions, where the dissertation was conducted. The PhD dissertation was carried out in the framework of the scientific projects of the Institute of Nuclear Physics, Scientific Laboratory of Theoretical Nuclear Physics "Development of highly effective variational methods for solving problems of quantum physics in several bodies" (2020-2022), UT-FA-2020-3 "Ultracold phase transitions in disordered quantum magnets and atomic gases with long range interactions" (2020-

2022);

The aim of the research is to develop Hartree-Fock-Bogoliubov approximation and Bose-Einstein condensate theory for anisotropic quantum magnets at very low temperatures as well as determine the phase transitionsins in these materials.

The tasks of the research:

to propose the thermodynamic potential of triplons at low temperature based on Hartree-Fock-Bogolyubov approximation including effect of anisotropy and anomalous density;

to determine quantum phase transitions in quantum magnets at a temperature close to zero;

to prove the existence of restrictions on magnetic phases and states of triplonic Bose-Einstein condensates;

to show the interference and Josephson transition of two Bose systems in quantum magnets at low temperature;

to obtain optimal values for fitting parameters of the exchange anisotropy and the Dzyaloshinskii-Moriya interaction within the mean field approach;

to describe existing experimental results on magnetization and the energy dispersion relation of quantum magnets with anisotropies, especially for the sample $TlCuCl_3$.

The objects of the research are antiferromagnetic materials, $TlCuC_3$ compound.

The subjects of the research are possible Bose-Einstein condensates of magnetic quasiparticles in quantum antiferromagnets at low temperatures and related problems.

The methods of the research is a new theoretical approach - the mean-field theory; bose-Einstein condensate theory; Hartree-Fock-Bogolyubov (HFB) approximation; second quantization method.

The scientific novelty of the research is the follows:

it was shown that using the mean-field theory for antiferromagnetic materials, i.e. anisotropic quantum magnets, the Hartree-Fock-Bogoliubov approximation was applicable for the regions below the critical temperature (T≤Tc);

it was firstly showen that taking into account the anisotropic properties of quantum magnets one can explain interference of two condensates and Josephson effects with the help of the phase difference of two matter waves;

it was firstly proven that consideration of Dzyaloshinskii-Moriya anisotropy, smears second order phase transition to a crossover, such that density of condensed particles diminishes asymptotically in the region of $T>T_c$ and the phase angle of condensate wave function may have only discrete values;

the experimental data on magnetization at low temperatures for anteferromagnetic material $TlCuCl_3$ was described by the help of Hartree-Fock-Bogoliubov approximation.

Practical results of the research are as follows:

for the first time, temperature dependence of the magnetization, heat capacity, and anomalous density of the condensate were calculated for quantum magnets and the results compared with the available experimental results for the $TlCuCl_3$ antiferromagnetic material;

for the first time, the restrictions placed on the phase of the condensate wave function have been studied in detail and it has been proved that the phase accepts only discrete values;

the transition from the ground state to the condensate state is shown to occur as crossover which is opposed to the second order phase transition;

using the results obtained for the phase the condensate wave function, the low-temperature phase transitions, Josephson effects and interference phenomena in quantum magnets are described.

The reliability of the research results is provided by the followings: modern methods of Bose-Einstein Condensation and the theoretical physics and highly effective numerical methods and algorithms are used; careful check of a consistence of the received theoretical results with experimental data and results of other authors is performed. Conclusions are well consistent with the main provisions of the field theory of quantum magnets and experimental results.

Scientific and practical significance of the research results. The scientific significance of the research results is determined by the ability of the developed description of the quantum critical behavior at the quantum phase transition via fundamental observables such as the specific heat, order parameter and field-induced magnetization. The comparison of the results of this dissertation and the observational data on antiferromagnetic materials can be explained well. We expect that our theoretical studies provide a new insight in understanding bose particle physics and BEC phenomena of magnons in quantum magnets.

Implementation of the research results. Based on the results obtained on the study of low temperature phase transitions in anisotropic quantum magnets:

the obtained results of Hartri-Fok-Bogolyubov's approximation for antiferromagnetic materials, ie anisotropic quantum magnets, has been used by foreign researchers (references in international journals: Physical Review B, 97, id.140405, 2018, Physical Review B, 98, id.144416, 2018, 34rd Youth Academic Annual Conference of Chinese Association of Automation, 18886028, 2019, International Journal of Modern Physics, 35, id.2150018, 2020, Physics Letters A, 384, id.126313 2020). The application of scientific results allowed the calculation of Tan’s contrast in antiferromagnets;

taking into account the anisotropic properties of quantum magnets, the observation of two condensate interference and the Josephson effect using phase difference of two matter waves has been used by foreign researchers (references in international journals: International Journal of Modern Physics B, 35, id.2150018, 2021, Journal of Physics: Condensed Matter, 33, id.465401, 2021). The application of scientific results has made it possible to develop a theoretical description of the phase angle;

considering the Dzyaloshinsky-Moria anisotropy, it was used by foreign researchers that the phase transition to the crossover, as well as the phase angle of the condensate wave function having only discrete values, were asymptotically decreases at T> Tc (references in international journals: International Journal of Modern Physics B, 35, id.2150018, 2021, Journal of Physics: Condensed Matter, 33, id.465401, 2021, Physics Letters A, 384, id.126313 2020). The application of scientific results allowed to analyze quantum phase transition in the quantum antiferromagnets ($CsFeCl_3$);

by the Hartree-Fock-Bogolyubov approximation, a detailed theoretical description of the experimental data for magnetization at low temperatures in anteferromagnetic material $TlCuCl_3$ has been used by foreign researchers (references in international journals: International Journal of Modern Physics B, 35, id.2150018, 2021, Journal of Physics: Condensed Matter, 33, id.465401, 2021). The application of scientific results has made it possible to determine the critical properties of Tans contact in bose systems.

Testing of the research results. The research results were reported and tested at 6 international and local scientific conferences.

Publication of the research results. On the theme of dissertation 10 scientific works were published, including 3 scientific papers in 3 international scientific journals recommended by the Supreme Attestation Commission of the Republic of Uzbekistan for publishing basic scientific results of PhD dissertations.

Volume and structure of the dissertation. The PhD dissertation consists of an introduction, three chapters, conclusion, appendix and a bibliography. The size of the dissertation is 103 pages.

List of publications:

1. Narzikulov Z., Khudoyberdiev A. Critical properties of optical bosonic gases in

cubic optical lattices at arbitrary integer fillings // Uzbek Journal of Physics. – Tashkent (Uzbekistan), 2016. –Vol. 18 (№4). – pp. 238-245. (01.00.00. №5)

2. Khudoyberdiev A., Rakhimov A., Schilling A. Bose-Einstein Condensation of triplons with a weekly broken U(1) symmetry // New Journal of Physics. – IOP Publishing (United Kingdom), 2017. – Vol. 19. – pp. 113 (№9. Journal Citation Reports; IF=3.729).
3. Rakhimov A., Khudoyberdiev A., Rani L., Tanatar B. Spin-gapped magnets with weak anisotropies I: Constraints on the phase of the condensate wave function //Annals of Physics. – Elsevier (Netherland), 2021. – Vol. 424. – id.168361 -23 p. (№3. Journal Citation Reports; IF=2.73).
4. Rakhimov A., Khudoyberdiev A., Tanatar B. Effects of exchange and weak Dzyaloshinsky-Moriya anisotropies on thermodynamic characteristics of spin-gapped magnets // International Journal of Modern Physics B. – World Scientific(Singapore), 2021. –Vol.35. – id.2150223. -23 p. (№2. Journal Citation Reports; IF=1.219).
5. Rakhimov A., Khudoyberdiev A. The Low Temperature properties of anisotropic quantum magnets with a spin gap // “International Confrence on Superconductivity and Magnetism”, April 29-May 04 2018. -Antalya (Turkey), 2018. – pp. 291.
6. Xudoyberdiev A. Triplonlarning dzyaloshinsky-moriya va almashinuv anizatropiyalari tasiridagi boze enshteyn kondensatsiyasi // "Yadro fizikasi va yadroviy texnologiyalar", O’zbekiston yosh fiziklari V respublika anjumani, 4-5 December 2018. – Tashkent: Yadro fizikasi instituti, 2018. –pp. 65-66.
7. Khudoyberdiev A. Bose-Einstein condensation of triplons with anisotropy of the system // “II International Scientific Forum Nuclear Science and technologies”, 24-27 June 2019. – Almaty (Kazakhstan), 2019 – pp. 136.
8. Khudoyberdiev A., Rakhimov A. Anisotropic properties of antiferromagnetic materials at low temperatures // International Conference "Fundamental and

applied problems of Physics", 22-23 September 2020. – Tashkent (Uzbekistan), 2020 – pp. 29.

9. Khudoyberdiev A., Narzikulov Z. The phase angle of the triplon condensate wave function with and without anisotropy // "Yadro fizikasi va yadroviy texnologiyalar", O’zbekiston yosh fiziklari VI respublika anjumani, 01-02 December 2020. – Tashkent: Institute of Nuclear Physics, 2020 – pp. 59-65.
10. Abdurakhmonov T., Khudoyberdiev A. Restrictions on the phase angle of the triplon gas wave function // “Yosh olimlar va fizik talabalarning I Respublika ilmiy anjumani”, 21 April 2021. –Tashkent (Uzbekistan), 2021. –pp. 243.

I. BOSE-EINSTEIN CONDENSATION IN QUANTUM MAGNETS WITH ANISOTROPY

§ 1.1. Introduction

There are many kinds of magnetic and non magnetic materials with different physical properties. Ferromagnets and antiferromagnets are more famous for their different properties in various conditions. Ferromagnetic materials have a high value of magnetic susceptibility and strong magnetization. If we apply an external magnetic field on ferromagnetic material, we obtain strong magnetization as neighboring spins of the atoms lie in a parallel form with the same direction. But, for antiferromagnetic materials, the scenario is different since each atom shows spin direction with antiparallel to neighbouring one. However, for antiferromagnetic materials, more interesting effects occur at low temperatures. Naturally, it is expected that at absolute zero temperature magnetization of antiferromagnetic materials should be zero. But, experimental observations for some compounds show that magnetization starts to increase as temperature decrease to near absolute zero and this effect occurs above some critical external magnetic field [1; pp. 13701-4]. Theoretically, there have been many attempts to explain this phenomena. One of the good ways is the Bose-Einstein Condensation of quasiparticles in these systems.

At early times it is believed that only gases with integer spin may be condensated as these states obtained for ^{4}He, which obeys Bose statistics [2; pp.110-135, 3; pp. 26-51, 4; pp. 569-582]. However, experimental results showed that not only gas and liquid but also quasiparticles in solids may go to the condensate at low temperature [1; pp. 13701-4]. For solids, such condensation occurs at higher temperatures compared to gas or liquid. At low temperature, in some antiferromagnetic materials, joined antiparallel spin systems with integer spin are called dimers and these dimers may go to the condensate by forming quasiparticles, elementary excitations, magnons. The density of magnons can be changed by an applied external magnetic field and they exhibit non-zero staggered magnetization at low temperature These types of antiferromagnetic materials are called quantum magnets or spin-gapped magnets as one can explain their effects by quantum field theory. In this work, we consider antiferromagnetic sample $TlCuCl_3$. This compound has a monoclinic crystal structure and dimer with spin S=1/2 from Cu^{2+} ions create elementary excitations, magnons [5; pp. 62-65].

The role of spontaneous symmetry breaking (SSB) is also essential in condensed matter and particle physics. In the standard model of particle physics, SSB of gauge symmetries is responsible for separating the weak and electromagnetic forces as well as creating several particles with mass [2; pp. 110-135]. SSB also lies in the basis of theories such as BEC, ferromagnetism, superconductivity, etc in condensed matter physics. Due to this symmetry breaking, the system Hamiltonian is invariant for the transformation of field operators, but its ground state does not become invariant. One can check this by substituting field operators as $\psi(\mathbf{r}) \rightarrow \psi(\mathbf{r})e^{i\alpha}$ while α in the exponent is a real number. It is also stated that SSB is a required condition to obtain BEC state [3; pp. 26-51]. For the solid system, this transition can be viewed as a BEC of elementary excitations, bosonic quasiparticles. Moreover, to write the Hamiltonian for solid state, one may consider these particles as real interacting magnons. Now, we have some space with an unknown number of magnons and it is interesting to study this system by quantum field theory. For the first time, this theory has been applied to describe the properties of superfluid helium for a lattice model, connecting it to the problem of a spin $1/2$ ferromagnet with anisotropic exchange coupling in an applied the external magnetic field. The appearance of a transverse spin ordering is then considered to be a manifestation of off-diagonal long-range order associated with the BEC of the real interacting particles [4; pp. 569-582]. In these systems, the peculiar properties of some antiferromagnets have also been successfully analyzed in terms of a BEC, namely that of interacting bosonic quasiparticles. At low temperatures, as we put quantum antiferromagnetic material into the external magnetic field, there occurs Zeeman effect and it modifies additional energy states. Quasiparticles can rejoin in three different energy state with S=-1, 0, +1 and these bosonic quasiparticles are called “triplons” as they lie into triple state. To obtain BEC of triplons one needs to increase external magnetic field above some critical value until S=+1 state crosses the ground state. As a result, condensation of triplons explains properties of antiferromagnetic materials belove critical temperature such as magnetization, heat capacity normal and anomalous densities.

Magnetization measurements were performed to find the critical behavior of the field-induced magnetic ordering in the quantum antiferromagnetic $TlCuCl_3$ in 1999 [1; pp. 265-271]. They found experimentally an unusual shape of the magnetization curve, i.e. $M(H,T)$ for the different amounts of the external magnetic field from 5 T to 7 T, by above its critical value. In fact, as it is can be seen from Figure 3 of Ref. [1; pp. 265-271] , for $H_{\text{ext}} >$5.3 T, there is a critical temperature T_c below which the magnetization of the antiferromagnet starts to increase. Later on, some other experimental works [5; pp. 62-65, 6; pp. 13701-4, 7; pp. 58-68] were also published, who found similar results as in Ref. [1; pp. 265-271]. According to these works, there are some general conclusions about quantum magnets. In some antiferromagnets such as $TlCuCl_3$ and $KCuCl_3$ two Cu^{2++} ions are antiferromagnetically coupled to form a dimer in a crystal knots: the dimer ground state is a spin singlet (S=0), separated by an energy gap from the first excited triplet state with S=+1. This gap can be closed by the external magnetic field above some critical value. The below critical temperature $T < T_c$, elementary excitations, i.e. triplons, can undergo BEC. The number of triplons can be changed by the external magnetic field and which is taken as a chemical potential of the system. Total density of triplons ρ and the fraction of condensed triplons, ρ_0, defines the $M_{\parallel}$ and $M_{\perp}$ magnetizations as $M_{\parallel} \sim \rho$ and $M_{\perp} \sim \sqrt{\rho_0}$.

The properties of these quantum magnets with real gas of bosons can be studied and analyzed by the following effective Hamiltonian

$$H = \sum_{\mathbf{k}} (\varepsilon_k - \mu) a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + \frac{1}{2} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} U_{\mathbf{q}} a_{\mathbf{k}+\mathbf{q}}^{\dagger} a_{\mathbf{k}'-\mathbf{q}}^{\dagger} a_{\mathbf{k}} a_{\mathbf{k}'} \quad (1.1)$$

where $\varepsilon_{\mathbf{k}}$ is kinetic energy, μ is the chemical potential written by the following formula

$$\mu = g\mu_B (H_{\text{ext}} - H_c), \quad (1.2)$$

and $a_{\mathbf{k}}(a_{\mathbf{k}}^{\dagger})$ are annihilation (creation) operators for a magnons with momentum **k**. In this work, we limit our case and interactions between triplons does not depend on distance. Hence, contact interaction type U_q can be taken as a constant U potential [8; pp. 27-62]. The amount of this interaction potential can be found experimentally and can be predicted theoretically as a fitting parameter.

Also, theoretically, the condensation of triplons is studied in other antiferromagnetic compounds and have been published [10; pp. 565-616]. Usually, it is assumed to diagonalize this Hamiltonian by Bogoliubov transformations, to obtain energy dispersion relation of quasiparticels. However, it is essential that the spectrum of collective exitations, E_k should be gapples and satisfy the Goldstone theorem [5; pp. 62-65].

$$\lim_{\mathbf{k}\to\mathbf{k}_0} E_k \sim c|\mathbf{k} - \mathbf{k}_0|, \quad \mathrm{Re}(\mathrm{E_k}) \geq 0, \ \mathrm{Im}(\mathrm{E_k}) \leq 0 \quad (1.3)$$

where $\mathbf{k}_0$ is a microscopically occupied single state and c is the sound velocity. Hence, one may conclude that magnetization data and energy spectrum for $TlCuCl_3$ can be well explained by BEC of triplons [5; pp. 62-65. 6; pp. 13701-4. 9; pp. 1145-1168. 10; pp. 565-616].

Additionally, as our sample is antiferromagnetic and the crystal structure is monoclinic, one needs to consider anisotropy of the system also for theoretical approaches. The Hamiltonian of the system should be different from the Hamiltonian of real interacting bose gases. In fact, electron spin resonance (ESR) [11; pp. 054431-5] and inelastic neutron scattering (INS) [12; pp. 267201-4] experiments on quantum antiferromagnets show the existance of anisotropy of the spectrum of magnetic excitations[2]. The anisotropy may be very essential around critical point for BEC [13; pp. 198-204] and the energy gap between triplet and singlet states can not be fully matched by Zeeman effect. One should state that the anisotropy is not taken into account in (1.1). For these reasons, it is suggested to consider additional anisotropic terms into the Hamiltonian (1.1) as follows

$$H_{\mathrm{DM}} = i\gamma' \int d^3r[\psi(\mathbf{r}) - \psi^{\dagger}(\mathbf{r})] \tag{1.4}$$

and

$$H_{\mathrm{EA}} = \frac{\gamma}{2}\int d^3r[\psi(\mathbf{r})\psi(\mathbf{r}) + \psi^{\dagger}(\mathbf{r})\psi^{\dagger}(\mathbf{r})]. \tag{1.5}$$

The anisotropic Hamiltonians (1.4) and (1.5) are called as Dzyaloshinsky-Moriya (DM) and exchange anisotropy (EA) interactions, respectively[3], γ' and γ are interaction parameters, correspondingly.

The role of anisotropy has been also studied before by Mean-Field Approximation methods [14; pp. 014438-6, 15; pp. 275-281]. In [15; pp. 275-281], authors used Hartree-Fock-Popov (HFP) approximation to explain magnetizations $M(T,H)$ and phase transitions in $TlCuCl_3$ and they have some conclusions. According to their results, the EA describes well condensate density but fails to explain experimental data. On the other hand, the DM interaction smears out the phase transition into a crossover, i.e. there is no critical temperature above which the condensed density vanishes.

[2] Table1 in reference. [10; pp. 565-616]

[3] See Appendix B for some details

However, it can explain only the experimental data on $M(T,H)$ for $H \parallel b$, but fails to accurately reproduce the data on $H \perp (1,0,\bar{2})$ and stuggered magnetization. We show that these problems may be solved by taking into account DM interaction term more accurately.

Thus, a full theoretical description of experimental magnetization data of $TlCuCl_3$, with the phase diagram, i.e. $T_c(H)$, is still missing [10; pp. 565-616], and a more sophisticated analysis beyond the HFP approximation is required for a better agreement with the experimental data. In Refs. [7; pp. 58-68, 15; pp. 275-281, 16; pp. 020407-4. 17; pp. 2499-2516] , it was shown that the above missing parts are due to the shortcomings of HFP approximation. To overcome these problems, one needs to use a more accurate approximation, e.g. Hartee-Fock-Bogolyubov (HFB).

In the present chapter, we propose an alternative MFA approach, HFB approximation, which gives a better description of the magnetization data on $TlCuCl_3$ including only the EA Hamiltonian H_{EA} (1.5) by using only three fitting parameters. With this Hamiltonian we shall consider intra dimer and inter dimer interactions in the monoclinic crystal of $TlCuCl_3$ [Appendix B].

It is well known that the main difference between HFP and HFB approximation lies in consideration of the anomalous density-σ, which is completely neglected in the HFP but can be taken into account in the HFB approximation [19; pp. 291-359, 20; pp. 489-506, 21; pp. 063612-9]. In our approximation, we assume that our theory should coincide with that of Sirker *et al.* [15; pp. 275-281] in the particular case when σ is set to zero. We will show that in the system with a weakly explicitly broken U(1) symmetry the anomalous density σ may survive even at $T > T_c$ in contrast to the case with the SSB. Assuming that our theory must coincide in general with the HFB approximation of Ref. [21; pp. 063612-9], when $\gamma \to 0$, we shall extend this method to the case of a weak anisotropy.

This Chapter I is organized as follows. Firstly, we present our total Hamiltonian for the system. Then we will analyze U(1) symmetry breaking and its effects. Next, we apply our approximation for antiferromagnetic material $TlCuCl_3$ and show that our theory gives better description of magnetization curves. In the last section, we summarizes our results and conclusion.

§ 1.2. The Hamiltonian for interacting triplons with EA

As we outlined in the introduction, we consider only interaction within and neighbouring dimers in crystal. Then, the Hamiltonian for elementary excitations, interacting bose type particles (triplons, created by dimers) may be written in the following form

$$H = \int d^3r[\psi^\dagger(\mathbf{r})(\widehat{K} - \mu)\psi(\mathbf{r}) + \frac{U}{2}\left(\psi^\dagger(\mathbf{r})\psi(\mathbf{r})\right)^2 + \frac{\gamma}{2}\left(\psi^\dagger(\mathbf{r})\psi^\dagger(\mathbf{r}) + \psi(\mathbf{r})\psi(\mathbf{r})\right)] \tag{1.6}$$

where $\psi(\mathbf{r})$ is the field operator, U is the interaction potential and $\widehat{K}$ is the kinetic energy operator which defines the bare triplon dispersion ε_k in momentum space. The integration is performed over the unit cell of the crystal with the corresponding momenta defined in the first Brillouin zone. The parameter μ characterizes an additional direct contribution to the triplon energy due to the external magnetic field H_{ext},

$$\mu = g\mu_B H_{\text{ext}} - \Delta_{\text{st}} \tag{1.7}$$

and can be considered as a chemical potential of the $S_z = +1$ triplons. In Eqs. (1.2) and (1.7) g is the electron Landé factor and Δ_{st} is a spin gap separating the singlet ground state from the lowest-energy triplet excitations, $\Delta_{st} = g\mu_B H_c$, where H_c is the critical field when the triplons start to form.

We assume that the EA is described by the last term in (1.6). It is clear that this term violates U(1) symmetry, $\psi(\mathbf{r}) \to e^{i\varphi}\psi(\mathbf{r})$ explicitly, so strictly speaking there would be neither a Goldstone mode nor a Bose condensation [23; pp. 460-513]. For the occurance of BEC, we need to divide field operators into two parts by Bogolyubov shift

$$\psi(r) = \phi_0(r) + \tilde{\psi}(r), \quad \psi^{\dagger}(r) = \phi_0(r) + \tilde{\psi}^{\dagger}(r), \tag{1.8}$$

Here, $\phi_0(\mathbf{r})$ and $\tilde{\psi}(\mathbf{r})$ belong to the density of condensed and uncondensed particles, respectively. We also keep in mind the orthogonality of these functions $\phi_0(\mathbf{r})$ and $\tilde{\psi}(\mathbf{r})$, i.e

$$\int d^3r \tilde{\psi}(r)\phi_0(r) = 0 \tag{1.9}$$

and $\rho_0 = \phi_0^2$ is the density of condensed particles [23; pp. 460-513]. And, also $\rho_1 = (1/V)\int d^3r\langle\tilde{\psi}^{\dagger}(\mathbf{r})\tilde{\psi}(\mathbf{r})\rangle$ is the density of uncondensed particles, so that the total number of particles is

$$N = \int d^3r\langle\psi^{\dagger}(r)\psi(r)\rangle \tag{1.10}$$

In our case, this number defines the total density of triplons $\rho = N/V = \rho_0 + \rho_1$. At low temperatures there are two types of magnetization for antiferromagnetic materials. One of them is the total magnetization which is proportional to the total density of triplons $M = g\mu_B\rho$ and the second one is the staggered magnetization given by $M_{\perp} = g\mu_B\sqrt{\rho_0/2}$. This staggered magnetization is explained by BEC of triplons and occurs below the critical temperature. In this Chapter, we assume that there is a critical temperature that can be found as $\rho_0(T_c) = 0$ and $\rho_0(T \geq T_c) = 0$. Then, above T_c the total density of particles is defined as the density of non-condensed particles $\rho(T \geq T_c) = \rho_1$. However, because of anisotropy, the energy spectrum has a gap in the whole temperature regime.

We use HFB approximation to diagonalize Hamiltonian (1.6) [17; pp. 2499-2516]. As there is some quantum fluctuations, we apply Fourier transformation for the field operators

$$\tilde{\psi}(\mathbf{r}) = \sum_{\mathbf{k}} a_{\mathbf{k}} e^{i\mathbf{kr}} \ , \quad \tilde{\psi}^{\dagger}(\mathbf{r}) = \sum_{\mathbf{k}} a_{\mathbf{k}}{}^{\dagger} e^{-i\mathbf{kr}} \tag{1.11}$$

In this transformation, summation is $\sum_{\mathbf{k}} \rightarrow \frac{1}{(2\pi)^3}\int d\vec{k}_x d\vec{k}_y d\vec{k}_z$. Additionally, in our case, this integral should consider anisotropic property of the system. For convenience, we give a brief discription of this integral in Appendix A.

In the Hamiltonian (1.6), we consider Bogoliubov shift (1.8) and Fourier transformation. As a result, we divide it into five terms according to degree of field operators and with respect to annihilation and creation operators

$$H = H_0 + H_1 + H_2 + H_3 + H_4 \tag{1.12}$$

Here, the first term H_0 does not include field operators and gets the following form

$$H_0 = -\mu\phi_0^2 + \gamma\phi_0^2 + \frac{U}{2}\phi_0^4. \tag{1.13}$$

The next term with the first degree of filed operators

$$H_1 = \sum_{\mathbf{k}} \{a_{\mathbf{k}}^{\dagger}\sqrt{\rho_0}(\gamma - \mu + U\rho_0) + a_{\mathbf{k}}\sqrt{\rho_0}(\gamma - \mu + U\rho_0)\}\delta_{\mathbf{k},0} \tag{1.14}$$

Our main Hamiltonian with the second degree of filed operators, have the form

$$H_2 = \sum_{\mathbf{k}} (\varepsilon_k - \mu + 2U\rho_0)a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}} + \frac{U}{2}(\gamma/U + \rho_0)\sum_{\mathbf{k}} (a_{\mathbf{k}}a_{-\mathbf{k}} + a_{\mathbf{k}}^{\dagger}a_{-\mathbf{k}}^{\dagger}) \tag{1.15}$$

For the last two terms of (1.12), we have the formulas below

$$\begin{aligned} H_3 &= U\sqrt{\rho_0}\sum_{\mathbf{k,p}} [a_{\mathbf{p}}^{\dagger}a_{\mathbf{p-k}}a_{\mathbf{k}} + a_{\mathbf{k}}^{\dagger}a_{\mathbf{p-k}}^{\dagger}a_{\mathbf{p}}], \\ H_4 &= \frac{U}{2}\sum_{\mathbf{k,p,q}} a_{\mathbf{k}}^{\dagger}a_{\mathbf{p}}^{\dagger}a_{\mathbf{q}}a_{\mathbf{k+p-q}} \quad . \end{aligned} \tag{1.16}$$

In order to diagonalize our Hamiltonian with second, third and forth orders we use the

Wick theorem. This theory helps us to lower the third and fourth orders and add them to the second order Hamiltonian

$$a_{\mathbf{k}}^{\dagger}a_{\mathbf{p}}a_{\mathbf{q}} \rightarrow 2\langle a_{\mathbf{k}}^{\dagger}a_{\mathbf{p}}\rangle a_{\mathbf{q}} + a_{\mathbf{k}}^{\dagger}\langle a_{\mathbf{p}}a_{\mathbf{q}}\rangle,$$
$$a_{\mathbf{k}}^{\dagger}a_{\mathbf{p}}^{\dagger}a_{\mathbf{q}}a_{\mathbf{m}} \rightarrow 4a_{\mathbf{k}}^{\dagger}a_{\mathbf{m}}\langle a_{\mathbf{p}}^{\dagger}a_{\mathbf{q}}\rangle + a_{\mathbf{q}}a_{\mathbf{m}}\langle a_{\mathbf{k}}^{\dagger}a_{\mathbf{p}}^{\dagger}\rangle + a_{\mathbf{k}}^{\dagger}a_{\mathbf{p}}^{\dagger}\langle a_{\mathbf{q}}a_{\mathbf{m}}\rangle - 2\rho_1^2 - \sigma^2, \tag{1.17}$$

Here, multiplication of creation and annihilation operators with its average is proportional to the number of triplons as $\langle a_{\mathbf{k}}^{\dagger}a_{\mathbf{p}}\rangle = \delta_{\mathbf{k},\mathbf{p}}n_{\mathbf{k}}$, while multiplication of annihilation (creation) operators with its average is responsible for anomalous density, results of pair-correlated triplons $\langle a_{\mathbf{k}}a_{\mathbf{p}}\rangle = \delta_{\mathbf{k},-\mathbf{p}}\sigma_{\mathbf{k}}$. Hence, in these notations $n_{\mathbf{k}}$ and $\sigma_{\mathbf{k}}$ give the normal (ρ_1) and anomalous (σ) densities as following

$$\rho_1 = \textstyle\sum_{\mathbf{k}} n_{\mathbf{k}} = \sum_{\mathbf{k}} \langle a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}\rangle, \tag{1.18}$$

$$\sigma = \textstyle\sum_{\mathbf{k}} \sigma_{\mathbf{k}} = \frac{1}{2}\sum_{\mathbf{k}} \left(\langle a_{\mathbf{k}}a_{-\mathbf{k}}\rangle + \langle a_{\mathbf{k}}^{\dagger}a_{-\mathbf{k}}^{\dagger}\rangle\right). \tag{1.19}$$

Here, we restate that the difference between the HFB and HFP methods due to the anomalous density: by omitting σ and $\langle a_k a_p\rangle$ in (1.17) one may use the HFP approximation, which can also be obtained in variational perturbation theory [24; pp. 214-230]. However, in HFB theory, σ is accounted for an important term and it can not be neglected without making the theory not self-consistent [25; pp. 599-639. 26; pp. 506-511. 27; pp. 264-270]. Additionally σ has a dependence on temperature and external magnetic field. The application of HFB approximation and Wick's theorem give the following Hamiltonian which is divided into three parts

$$H = H_0 + H_{\mathrm{lin}} + H_{\mathrm{bilin}}, \tag{1.20}$$

$$H_0 = -\mu\rho + \gamma\rho_0 + \frac{U}{2}\rho_0^2 - \frac{U}{2}(2\rho_1^2 + \sigma^2), \tag{1.21}$$

$$H_{\text{lin}} = \sqrt{\rho_0}\sum_{\mathbf{k}} \{a_{\mathbf{k}}^{\dagger}[\gamma - \mu + \rho_0 U + 2\rho_1 U + \sigma U] + a_{\mathbf{k}}[\gamma - \mu + \rho_0 U + 2\rho_1 U + \sigma U]\}, \tag{1.22}$$

$$H_{\text{bilin}} = \sum_{\mathbf{k}} (\varepsilon_k - \mu + 2U\rho)a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}} + \frac{U(\gamma/\mathrm{U} + \rho_0 + \sigma)}{2}\sum_{\mathbf{k}} (a_{\mathbf{k}}a_{-\mathbf{k}} + a_{\mathbf{k}}^{\dagger}a_{-\mathbf{k}}^{\dagger}). \tag{1.23}$$

From (1.22), it is usually required that $H_{\text{lin}} = 0$ [28; pp. 235-272]. As a result, we get an equation for chemical potential:

$$\mu = U[\rho_0 + 2\rho_1 + \sigma + \gamma/\mathrm{U}]. \tag{1.24}$$

This relation is also the need of Hugenholtz and Pines theorem. The equation (1.24) can be obtained by minimization of the thermodynamic potential $\partial\Omega/\partial\rho_0 = 0$ [29; pp. 063602 – 11]. Now, our task is to diagonalize the Hamiltonian (1.23) and find the energy of the system by its average. We give these calculations in the Appendix B. As a result of diagonalization, we get the quasiparticle dispersion relation and densities for triplons

$$E_k = \sqrt{(\varepsilon_k + X_1)(\varepsilon_k + X_2)}. \tag{1.25}$$

$$X_1 = -\mu + U[2\rho + \tilde{\gamma} + \rho_0 + \sigma] \tag{1.26}$$

$$X_2 = -\mu + U[2\rho - \tilde{\gamma} - \rho_0 - \sigma]. \tag{1.27}$$

$$\Sigma_n = 2U\rho = (X_1 + X_2)/2 + \mu, \qquad \Sigma_{an} = U[\sigma + \rho_0 + \tilde{\gamma}] = (X_1 - X_2)/2. \tag{1.28}$$

Additionally, the density of uncondensed particles and anomalous density have the following form

$$\rho_1 = \frac{1}{V}\sum_{\mathbf{k}} \left\{\frac{W_k[\varepsilon_k+(X_1+X_2)/2]}{E_k} - \frac{1}{2}\right\}, \tag{1.29}$$

$$\sigma = \frac{X_2-X_1}{2V}\sum_{\mathbf{k}} \frac{W_k}{E_k}, \tag{1.30}$$

where $W_k = \coth(E_k/2\mathrm{T})/2 = f_B(E_k) + 1/2,\ f_B(E_k) = 1/(e^{E_k/T} - 1)$. From (1.29) and (1.30), one can see that they have the dependence on temperature. It means, the densities change at low temperature and can explain the properties of BEC of triplons.

§ 1.3. Spontaneous Breaking of Symmetry case

It is expected that the normal and anomal self- energies (1.28) should satisfy the Hugenholtz-Pines theorem [20; pp. 489-506]:

$$\Sigma_n - \Sigma_{an} = \mu. \tag{1.31}$$

However, equations (1.28) gives the result

$$\Sigma_n - \Sigma_{an} = X_2 + \mu. \tag{1.32}$$

So, the Hugenholtz-Pines theorem is satisfied when $X_2 = 0$. And also, this gives the gapless energy dispersion

$$E_k|_{X_2=0} = \sqrt{(\varepsilon_k + X_1)\varepsilon_k} = ck + O(k^3). \tag{1.33}$$

On the other hand, we may set in (1.27) as $X_2 = 0$ and $\gamma = 0$ to get

$$\mu = U[2\rho - \rho_0 - \sigma] = U[2\rho_1 + \rho_0 - \sigma]. \tag{1.34}$$

Interestingly, we get two different equations for chemical potential with (1.24) and above (1.34) formulas. The chemical potentials are the same in HFP approximation when $\sigma = 0$. However, when σ is taken into account i.e. when one is dealing with the HFB approximation , they are different. Moreover, in the SSB phase, Hugenholtz-Pines theorem is consistent in the HFP but not in the HFB approximations. To overcome this unclear difference, the solution was proposed in Ref. [21; pp. 063612-9]. Clearly, they suggested taking two different chemical potentials, μ_0 and μ_1. Then, μ_0 is responsible to satisfy the stability condition $H_{\text{lin}} = 0$ while μ_1 satisfies the requirement of gapless of the spectrum [21; pp. 063612-9, 30; pp. 349-399]. Hence, we have the following chemical potentials:

$$\mu_0 = U[2\rho_1 + \rho_0 + \sigma], \tag{1.35}$$

$$\mu_1 = U[2\rho_1 + \rho_0 - \sigma], \tag{1.36}$$

The total chemical potential

$$\mu = (\mu_0\rho_0 + \mu_1\rho_1)/\rho, \tag{1.37}$$

so that $N_0 = -(\partial\Omega/\partial\mu_0)_V$ and $N_1 = -(\partial\Omega/\partial\mu_1)_V$. Fortunately, above the critical temperature $\rho_0 = 0$, $\rho = \rho_1$ and $\mu = \mu_1$.

Now, we rewrite the equations (1.24), (1.26) and (1.27) with two differenet chemical potentials

$$\mu_0 = U[2\rho_1 + \rho_0 + \sigma + \gamma/U)], \tag{1.38}$$

$$X_1 = -\mu_1 + U[2\rho + \gamma/U + \rho_0 + \sigma], \tag{1.39}$$

$$X_2 = -\mu_1 + U[2\rho - \gamma/U - \rho_0 - \sigma]. \tag{1.40}$$

However, the equations (1.25) , (1.29) and (1.30) are kept without a change.

§ 1.3.1. Condensed phase where $T \le T_c$

We take into account that in (1.29) and (1.30) formulas

$$\rho_1 = \rho_1(X_1, X_2) \qquad \sigma = \sigma(X_1, X_2) \tag{1.41}$$

This means that we have three equations with respect to four unknown quantities: X_1, X_2, μ_0 and ρ_0. If we include the anisotropy, the Hugenholtz -Pines theorem will not be satisfied. Fortunately, the anisotropic parameter γ is rather small. For this reason, we state that the Hugenholtz -Pines theorem may be violated up to first order in anisotropic parameter by neglecting higher order terms $O(\gamma^2)$. Thus, we assume that

$$\Sigma_n - \Sigma_{an} = \mu_1 + 2\gamma c_\gamma, \tag{1.42}$$

where c_γ is a real number. Now, we can get following equation for X_1, X_2 and μ_1

$$X_2 = \Sigma_n - \Sigma_{an} - \mu_1 = 2\gamma c_\gamma. \tag{1.43}$$

$$\mu_1 = U[\rho_0 + 2\rho_1 - \sigma - \gamma(1 + 2c_\gamma)/\mathrm{U}], \tag{1.44}$$

$$X_1 = 2U[\rho_0 + \sigma + \gamma(1 + c_\gamma)/\mathrm{U}], \tag{1.45}$$

Now, as expected, our energy spectrum has a gap due to the anisotropy

$$E_k = \sqrt{(\varepsilon_k + X_1)(\varepsilon_k + 2\gamma c_\gamma)}, \quad E_k|_{k\to 0} = \sqrt{2X_1\gamma c_\gamma}. \tag{1.46}$$

In our case, μ_1 is external parameters given by external magnetic field. From (1.44) and (1.45) we get the following equation

$$\Delta_1 \equiv \frac{X_1}{2} = \mu_1 + 2U(\sigma - \rho_1) + \gamma(2 + 3c_\gamma), \tag{1.47}$$

We use dimensionless terms as $Z = \Delta_1/\mu_1$ and taking into account (1.29) , (1.30) we obtain

$$Z = 1 + \tilde{\sigma} - \tilde{\rho}_1 + \frac{\gamma}{2U\rho_c^0}(2 + 3c_\gamma), \tag{1.48}$$

$$\tilde{\rho}_1 \equiv \tilde{\rho}_1(Z) = \frac{\rho_1(Z)}{\rho_c^0} = \frac{1}{\rho_c^0}\sum_{\mathbf{k}} \left\{\frac{W_k(\varepsilon_k + \Delta_1 + \Delta_2)}{E_k} - \frac{1}{2}\right\}, \tag{1.49}$$

$$\tilde{\sigma} \equiv \tilde{\sigma}(Z) = \frac{\sigma(Z)}{\rho_c^0} = \frac{\Delta_2 - \Delta_1}{\rho_c^0}\sum_{\mathbf{k}} \frac{W_k}{E_k}, \tag{1.50}$$

where $\rho_c^0 = \mu_1/2U$, $\mu_1 = g\mu_B(H_{\text{ext}} - H_c)$, $E_k = \sqrt{(\varepsilon_k + 2\Delta_1)(\varepsilon_k + 2\Delta_2)}$, and $\Delta_2 = \gamma c_\gamma$.

In this phase, we solve (1.48)-(1.50) equations with respect to Z. And denoting the solution as Z_0, we get the following result

$$\rho_0 = 2Z_0\rho_c^0 - \sigma(Z_0) - \gamma(1 + c_\gamma)/\mathrm{U} \tag{1.51}$$

This ρ_0 is the density of condensed elementary excitations (triplons). The total number of particles may be found from $\rho = \rho_0 + \rho_1(Z_0)$ where $\rho_1(Z_0)$ is evaluated by equation (1.49) .

§ 1.4. Magnon fraction and the critical temperature

For a given external magnetic field, as we increase the temperature up to the critical point the density of condensed triplons will decrease and there should be a critical point, $T = T_c$, $\rho_0(T \geq T_c) = 0$. The amount of density in this point is considered as critical density $\rho(T_c) = \rho_c$. So, our task is to find these quantities from the above equations. Near the critical point, we have

$$\mu_1(T \to T_c) = U[2\rho_c - \sigma_c - \gamma(1 + 2c_\gamma)/U] = g\mu_B H_{\text{ext}} - \Delta_{\text{st}}. \quad (1.52)$$

$$\rho_c = \frac{g\mu_B H_{\text{ext}} - \Delta_{\text{st}}}{2U} + \frac{\sigma_c + \gamma(1+2c_\gamma)/\text{U}}{2} \equiv \rho_c^0 + \frac{\sigma_c + \gamma(1+2c_\gamma)/\text{U}}{2}. \quad (1.53)$$

The energy dispersion of quasiparticles have the form

$$E_k^c = E_k(T \to T_c) = \sqrt{(\varepsilon_k + X_1^c)(\varepsilon_k + 2\gamma c_\gamma)}, \quad (1.54)$$

where X_1^c, by using (1.45) , is given as

$$X_1^c = X_1|_{T \to T_c} = 2U[\sigma_c + \gamma(1 + c_\gamma)/\text{U}]. \quad (1.55)$$

By the help of equations (1.29), (1.30) and (1.53) we can obtain two formulas for T_c and σ_c

$$\sum_{\mathbf{k}} \frac{f_B(E_k^c)}{E_k^c} [\varepsilon_k + U(\sigma_c + \gamma(1 + 2c_\gamma)/\text{U})] = \frac{g\mu_B H_{\text{ext}} - \Delta_{\text{st}}}{2U} + \frac{\sigma_c + \gamma(1+2c_\gamma)/\text{U}}{2} \quad (1.56)$$

$$\sigma_c = -U(\sigma_c + \gamma/U)\sum_{\mathbf{k}} \frac{f_B(E_k^c)}{E_k^c} \tag{1.57}$$

where $f_B(E_k^c) = 1/(\exp(E_k^c/T_c) - 1)$. We solve these equations by considering ρ_c^0 as the critical density at $\gamma = 0$, i.e. $\rho_c^0 = \rho_c(\gamma = 0) = \mu_1/2U$.

§ 1.4.1. Spontaneous Breaking of Symmetry phase without anisotropy

In this case, we should omit the anisotropic parametr γ. And we analyze the anomalous density above and belove the critical temperature. According to equation (1.57) with γ=0 we get

$$\sigma_c = -U\sigma_c \sum_{\mathbf{k}} \frac{f_B(E_k^c)}{E_k^c} \equiv -\sigma_c A_c, \tag{1.58}$$

where

$$A_c = U\sum_{\mathbf{k}} \frac{f_B(E_k^c)}{E_k^c}. \tag{1.59}$$

We know that interaction potential $U > 0$. Hence, the only possible solution of the equation (1.58) is:

$$\sigma_c|_{\gamma=0} = 0 \tag{1.60}$$

In the isotropic case, we can find the critical temperature T_c^0 by solving the equation (1.53).

$$\rho_c^0 = \frac{\mu}{2U} = \sum_{\mathbf{k}} \frac{1}{e^{\varepsilon_k/T_c^0} - 1} = \frac{g\mu_B H_{\text{ext}} - \Delta_{\text{st}}}{2U}, \tag{1.61}$$

§ 1.4.2. Anisotropic and normal phase where $T > T_c$

In this phase, we analyze anomalous density for $\gamma \neq 0$ case, for the whole range of temperature and density of triplons above the critical temperature. Now equation (1.58) has a formal solution for σ_c

$$\sigma_c = -\frac{\gamma A_c}{(1+A_c)U}, \tag{1.62}$$

It is expected that anomalous density of triplon is negative term [31; pp. 9341- 7]. Here, we also use dimensionless variables as $\sigma_c = -\gamma\sigma_x/U$ and $\rho_c = \rho_x\rho_c^0$ where now ρ_x and σ_x are just numbers.

The equations (1.56) and (1.61) are used to calculate the shift of critical temperature due to the effect of anisotropy which can be written as

$$\frac{\Delta T_c}{T_c^0} \equiv \frac{T_c - T_c^0}{T_c^0} \tag{1.63}$$

where $T_c^0 = T_c(\gamma = 0)$ is defined by equation (1.61) .

In the normal phase, $\rho_0 = 0$, $\rho_1 = \rho$, $\mu_1 = \mu$ and $T > T_c$, then our main equation (1.26) takes the form

$$X_1 = -\mu + U[2\rho + \sigma + \gamma/U], \tag{1.64}$$

$$X_2 = -\mu + U[2\rho - \sigma - \gamma/U], \tag{1.65}$$

Additionally, the total density of triplons is found by density of uncondensed one as we are analyzing the region above the critical temperature

$$\rho = \rho_1 = \sum_{\mathbf{k}} \frac{(\varepsilon_k - \mu_{eff}^0)}{E_k} f_B(E_k), \tag{1.66}$$

where $\mu_{eff}^0 = \mu - 2U\rho$.

As a result, in this phase, it is required to solve two equations with respect to ρ_x and σ_x

$$\rho_x - \frac{1}{\rho_c^0} \sum_{\mathbf{k}} \frac{\varepsilon_k}{E_k} - 2A(\rho_x - 1) = 0, \tag{1.67}$$

$$\sigma_x - \frac{A}{1+A} = 0, \tag{1.68}$$

The results of these calculations and other physical properties of the system will be given in the next section.

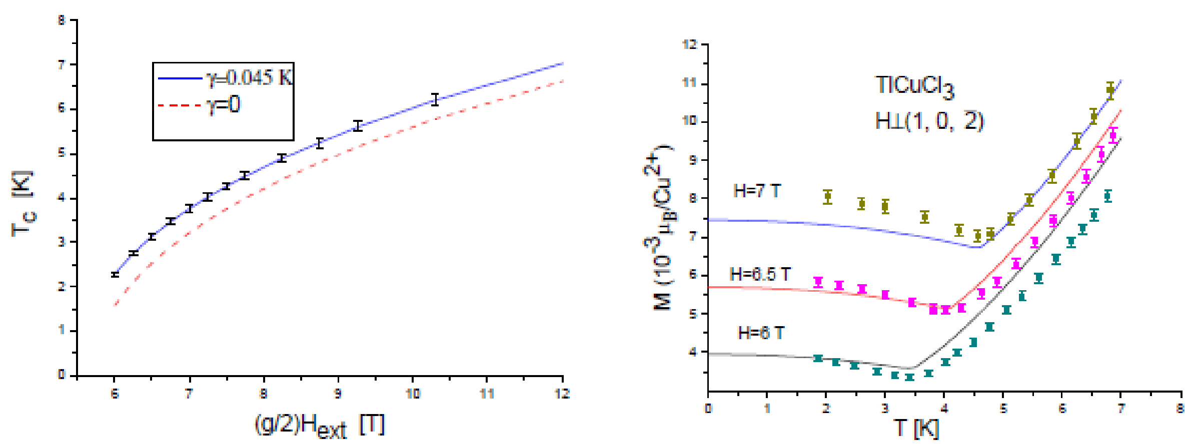
§ 1.5. Magnetization and critical temperature

The magnetization of quantum magnets well describes the BEC of triplons and the phase transition process. For this reason, at low temperatures, it is suggested to find two types of magnetizations in quantum magnets. They are total and staggered magnetizations that are proportional to the total and condensed density of triplons, respectively. One of the famous quantum magnets is $TlCuCl_3$ and there are many experimental results for this compound [4; pp. 265-271. 6; pp. 13701 – 4. 32; pp. 224403-8, 33; pp. 3429-3434] in the range of 5 T $\leq H_{\text{ext}} \leq$ 8 T, 2 K $\leq T <$ 7 K. Theoretically, the best way to explain these data is to include anisotropy in the crystal of the compound. Usually, theoretical works have difficulty while explaining staggered magnetization dependence on temperature data and this is explained as lack of HFP approximation. For this reason, we choose HFB approximation and try to apply our results for experimental data explanations with parameters γ, c_γ and U. Additionally, we use anisotropic bare dispersion [33; pp. 3429-3434] (see appendix A) rather than using quadratic or relativistic one [34; pp. 057201-4]. In Figs. 1.1 (a) and (b) we show a comparision between the experimental data and the resulting fits to these data. We have found that γ=0.045 K, $c_\gamma = 1.67$ and U=367.5 K. One should note that Lande factor has different value for different direction of external magnetic filed as $g = 2.23$ for $H_{ext} \perp (1,0,\bar{2})$ and $g = 2.06$ for $H_{ext} \parallel$ b. To explain phase diagram Fig.1.1 (a), there is a well-known formula as

$$T_c \sim (H_{\text{ext}} - H_c)^{\phi} \tag{1.69}$$

We have found that $\phi = 0.53$ (solid line) and $\phi = 0.62$ (dashed line) for the $\gamma = 0.045$ K and $\gamma = 0$ cases, respectively. This means that the inclusion of a finite exchange anisotropy reduces the exponent ϕ, and one does not need to expect $\phi = 2/3$ as it has been debated in the literature [33; pp. 3429-3434. 34; pp. 057201-4. 35; 020409-4. 36; pp. 061601- 4].

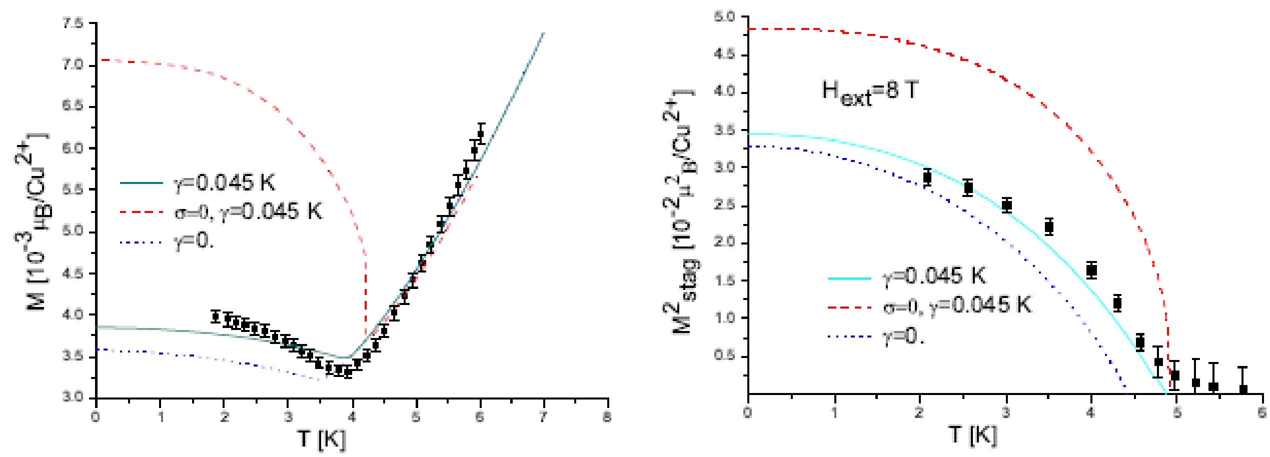
In Fig. 1.1(b) the magnetization curves for various H_{ext} are presented in comparison with the experimental data from Ref. [6; pp. 13701–4] for $H_{\text{ext}} \perp (1,0,\bar{2})$. It is seen that by taking into account the exchange anisotropy one can obtain an excellent agreement with the experimental results.



a) b)

Fig. 1.1. Phase diagram (a) and total magnetization (b) for $TlCuCl_3$. The experimental data are taken from [6; pp. 13701 – 4]

Using the above equations, we also obtain the total magnetization ($M = g\mu_B\rho$) for H ∥b, which is plotted in Fig.1.2(a) in comparison with the experimental data taken from [7; pp. 58-68] and with a corresponding calculations based on the HFP approximations [15; pp. 275-281]. It is seen that by neglecting the anomalous density σ (dashed line) or the exchange anisotropy γ (dotted line), one may reproduce the experimental magnetization only at high temperatures , $T > T_c$, while the inclusion of both, σ and γ makes it possible to obtain a significantly better theoretical description (solid line) for $T \leq T_c$ also. From the Fig. 1.2(a) one may conclude that the effect of the exchange anisotropy is rather large in the BEC - like phase and is almost negligible in the normal phase.



a) b)

Fig.1.2. The total magnetization (a) and square of the transverse magnetization (b) in different approximations for $H \parallel b$. The solid and dashed lines are for HFB and HFP approximations with the exchange anisotropy and the same input parameters, respectively. The dashed line in (a) is reproduced from Ref. [15; pp. 275-281] and the experimental data are taken from [7; pp. 58-68. 37; pp. 939-942]. The dotted line represents a HFB approximation without anisotropy, i.e. $\gamma = 0$, and $U = 367.5\text{K}$

Another important characteristics of quantum magnets is that the magnetically ordered state supports a staggered magnetization $M_\perp$, transverse to the field direction, leading to a canted antiferromagnetic state until the system becomes eventually fully polarized as the external field increases. In the BEC- scenario number of condensed particles is proportional to the square of the ordered transverse component:

$$M_{\perp} = g\mu_B\sqrt{\rho_0/2}, \tag{1.70}$$

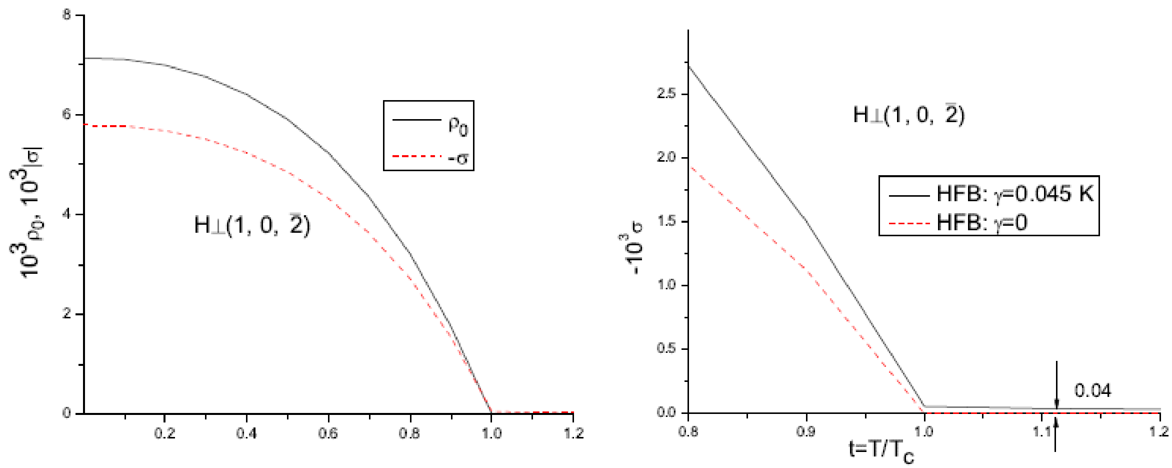
In our present approximation $M_{\perp}$ may be calculated directly from Eqs. (1.48) - (1.51) and (1.70) . The results are presented in Fig.1.2 (b), where we have used the same input parameters as in Figs.1.1 and experimental data taken from [37; pp. 939-942]. As it is seen from the figure the present approach with exchange anisotropy describes well the experimental data for $T \leq T_c$. A comparison of the dotted curve with the solid line in Fig. 1.2(b) shows that the exchange anisotropy enhances the staggered magnetization.

In present chapter, we have been only dealing with the exchange anisotropy, which gives a sharp phase transition with $\rho_0(T \geq T_c) = 0$. From magnetization graphs, it can be seen that our results best explain below the critical temperature. However, it is said that to describe the whole temperature regime, one needs to include DM anisotropy, so that $M_{\perp}^{\mathrm{DM}}(T \geq T_c) \neq 0$ [38; pp. 939-942. 39; pp. 184402- 8].

Finally, we discuss the role of the anomalous density whose absolute value is the density of pair correlated particles [40; pp. 96-110]. We present in Fig.1.3(a) the density of condensed particles ρ_0 (solid line) and the absolute value of the anomalous density $|\sigma|$ (dashed line) versus the reduced temperature. [4] It is seen that $|\sigma|$ is comparable with ρ_0 at all temperatures. Another interesting fact, which is demonstrated in Fig. 1.3(b) is that the anomalous density survives, although on a small level, even above the critical temperature where it vanishes asymptotically. In fact, this approach predicts the existence of two critical temperatures. T_c, where $\rho_0 = 0$, $\sigma \neq 0$ and another one T^*,

[4] Actually, $\sigma < 0$ in the whole region of temperatures.

where $\rho_0 = 0$, $\sigma = 0$, with $T^* > T_c$. This exotic state in the region $T_c < T < T^*$ has not been experimentally observed yet, but it is predicted to exhibit a modified dispersion relation.



a) b)

Fig.1.3. (a): The condensed (solid line) and the absolute value of anomalous densities (dashed line) in HFB approximation at $H_{\text{ext}} = 7$ T; (b): The behavior of $|\sigma(t)|$ near the critical temperature with (solid line) and without (dashed line) exchange anisotropy

§ 1.6. Conclusion for Chapter I

Assuming that the low temperature properties of quantum magnets with a weak U(1) symmetry breaking can be described in a BEC - like scenario, we proposed a new MFA based approach within the Hartree - Fock - Bogolyubov approximation, which takes into account an anomalous density σ and exchange anisotropy. This approach not only reproduces experimental data such as the critical temperature and the magnetization in a satisfactory way, but also removes certain inconsistencies and drawbacks met in the previously used Hartree - Fock - Popov approaches [15; pp. 275-281]. We have found $\gamma = 0.045$ K, $c_\gamma = 1.67$ and $U = 367.5$ K valid for both the

$H \parallel b$ and $H \perp (1,0,\bar{2})$ directions.

The present approach also gives a fair theoretical description of the staggered magnetization data for $T \leq T_c$ and predicts a plausible value for the critical exponent β. However, to improve the theoretical description of the experimental data on staggered magnetization the inclusion of Dzyaloshinsky-Moriya anisotropy also seems to be necessary. We have estimated the anisotropy - induced shift of the critical temperature, and show that it is substantial. Finally, we predict that the anomalous density is comparable to the condensed one, and survives at temperatures exceeding T_c where the condensate fraction is zero. The present approach has its intrinsic limits, however. It cannot describe the system below the critical magnetic field because there are no triplons at $H < H_{c_1}$. Moreover, it seems to fail to work at high field regime, where it may predict instability [16; pp. 020407-4] instead of the saturation of the magnetization due to the fully polarization of the spins. For more extended studies, it is required to differentiate the main difference between BEC of gases and BEC of triplons. One of the ways is to breafly analyze condensate wave function operator and find possible phases for this term. As a result, one can study additional phenomenas in quantum magnets such as superfluidity or Josephson effects [41; pp. 2301-2309]. Hence, our theoretical studies require to consider more realistic interaction between triplons and develop it by bond operator formalism [42; pp. 054423-20] or a another model proposed by Fischer *et al.* [43; pp. 47001 -7].

Represented results of calculation were published as scientific papers in:

- Khudoyberdiev A., Rakhimov A., Schilling A. Bose-Einstein Condensation of triplons with a weekly broken U(1) symmetry // New Journal of Physics– IOP Publishing (United Kingdom), 2017. – Vol. 19 – ip. 113002 – 18 p.
- Khudoyberdiev A. Bose-Einstein condensation of triplons with anisotropy of the system // “II International Scientific Forum Nuclear Science and technologies”, 24-27 June– Almaty (Kazakhstan), 2019 – pp. 136.

- A.Rakhimov, A.Khudoyberdiev. The Low Temperature properties of anisotropic quantum magnets with a spin gap // "International Confrence on Superconductivity and Magnetism", April 29-May 04–Antalya (Turkey), 2018– pp. 291.
- Z.Narzikulov, A.Khudoyberdiev. Critical properties of optical bosonic gases in cubic optical lattices at arbitrary integer fillings // Uzbek Journal of Physics – (Uzbekistan), 2016. –Vol. 18 (№4) – pp. 238-245.

II. Phase transition and restrictions on the phase angle of the condensate wave function

§ 2.1. Introduction

Historically, Bose-Einstein condensation was first observed in atomic gases and then in magnetic materials. Besides, in the last decade, it was reported that there is a class of atomic condensates, manifesting magnetic properties. These are referred to as spinor Bose-Einstein condensates. A spinor BEC consists of atoms with a spin degree of freedom [44; pp. 9323-9329]. In addition to exhibiting a spatial coherence, a spinor condensate also displays a range of spin orders, determined by the interactions and externally applied magnetic field. Various aspects of the phase diagram and condensate dynamics have been explored in the experiments, particularly for the case of spin-1 where the atoms can access three magnetic sublevels. An important feature of this system is that it exhibits a rich excitation spectrum with phonon and magnon branches.

Naturally, ultracold atoms with spin degrees of freedom have attracted considerable interest due to the possibility of modeling quantum magnetism and exploring the interplay between spatial and spin degrees of freedom [45; pp. 295-300]. These atoms can exhibit the properties of a ferromagnet or an antiferromagnet depending on the sign of the spin dependent interaction. Moreover, the competition between an external magnetic field and the ferromagnetic interaction gives rise to rich phases. For example, there would be two superfluid phases: the polar superfluid phase, in which only one component is superfluid, and the broken-axisymmetry (BA) superfluid phase, which has non-zero transverse magnetization. Further, a similar effect that could be related to triplon condensation was observed in other compounds and they are reviewed in Ref. [46; pp. 5816-5826]. Thus, dimerized quantum magnets present a compelling alternative environment due to an exact mapping to a lattice gas of bosons with hard-core repulsion. These systems consist of lattices of pairs of spins (dimers) which are all in a singlet configuration in the ground state. This state can be viewed as an "empty" lattice, where triplet excitations can be thought of as site occupied with S=1 bosons, i.e. triplons.

Mathematically, the phase χ and condensate fraction ρ_0 are given by the following representation of the field operator $\psi=\rho_0^{1/2}\chi + \psi_1$ where ψ_1 corresponds to quantum fluctuations. The choice of the phase by hand determines a physical state, e.g., $\chi=[1,0,0]$ corresponds to the ferromagnetic one [48; pp. 79-98]. The question arises, to what extent is this strategy justified? In other words, are there any physical constraints to the choice of the phase of a spinor BEC, dictated by requirements of stability? Looking for the answer to this question will be one of the tasks, to be considered in the present chapter.

In chapter I, we have shown that one needs to consider the effect of the anisotropies to study quantum magnets, theoretically. In general, the exchange interaction between two moments has the form $\Sigma_{ij} S_r^i T^{ij} S_{r+e_\nu}^j$ [47; pp. 180-401] where $\boldsymbol{T} = (1/3)Tr(\boldsymbol{T})I + \boldsymbol{T}_{as} + \boldsymbol{T}_{SM}$. Here the first term leads to the usual isotropic exchange coupling, $\boldsymbol{T}_{as}$ is an antisymmetric tensor that describes the Dzyaloshinsky-Moriya (DM) interaction $\boldsymbol{D} \cdot [\boldsymbol{S}_r \boldsymbol{S}_{r+e_\nu}]$, where $\boldsymbol{D}$ is the DM vector [49; pp. 129-134]. The last term contains the so-called symmetric exchange anisotropy (EA) and has contributions from the classical dipole-dipole interaction between magnetic moments. In this chapter, we study possible phases in quantum magnets by the inclusion of both interactions.

The phase of the condensate also plays an important role in interference experiments, where two BECs are released and resulting interference patterns are measured [54; pp. 637-641]. Hence, we also study the consequences of this conclusion for some physical phenomena such as interference of two Bose systems and Josephson effect.

As it is mentioned above that DM interactions best explain staggered magnetization and phase of the condensate wave function. That is why we study low temperature properties of spin gapped magnets with DM and exchange anisotropies ((1.4) and (1.5) Hamiltonians) by taking into account their contribution more systematically than in Refs. [15; pp. 275-281. 50; pp; 113002-18] including anomalous density. For this purpose, we shall represent DM and EA interactions by linear (in fields) and quadratic terms, respectively, and use δ-expansion method developed in quantum field theory [51; pp. 2710-2725. 52; pp. 014515-8. 53; 076001- 6]. Then, we calculate the thermodynamic potential of the system by path integral formalism and find some properties of the system, such as magnetization, heat capacity and energy dispersion relations.

§ 2.2. The thermodynamic potential including Exchange anisotropy and Dzyaloshinskii-Moriya interactions

To find the thermodynamic potential of triplon Bose gases, firstly we should write the Hamiltonian for the system. In this case, we propose that triplons interact in different ways as our system consists of solid material, i.e. antiferromagnetic monocline type crystal. Hence, we take into account isotropic and anisotropic interactions for effective Hamiltonian which can be written as follows

$$H = H_{iso} + H_{aniso}, \tag{2.1}$$

$$H_{iso} = \int d\vec{r} \left[\psi^{+}(r)(\widehat{K} - \mu)\psi(r) + \frac{U}{2}(\psi^{+}(r)\psi(r))^2\right], \tag{2.2}$$

$$H_{aniso} = H_{EA} + H_{DM}, \tag{2.3}$$

$$H_{EA} = \frac{\gamma}{2}\int d\vec{r}[\psi^{+}(r)\psi^{+}(r) + \psi(r)\psi(r)], \tag{2.4}$$

$$H_{DM} = i\gamma' \int d\vec{r}[\psi(r) - \psi^{+}(r)], \tag{2.5}$$

The linear Hamiltonian, in Eq. (2.5) corresponds to a simple case when singlet-triplet mixing is neglected and DM vector is chosen as $D \parallel x$ and $H \parallel z$. In the present work, we assume γ' to be small but finite to study its physical consequences [55; pp. 1892-1917]. A Similar form of the Hamiltonian can be found in Ref. [56; pp. 911-916]. Here, the Hamiltonian (2.2) has a symmetry for gauge transformation $\psi \to e^{i\phi}\psi$, while H_{aniso} has not. We assume γ and γ' to be small that one can deal with the occurance of Bose condensation. This corresponds to the macroscopic occupation of a single quantum state, from the remaining part of the Bose field operator. Therefore, assuming that just one state of the system to be occupied macroscopically, it is natural to re-arrange the Bose field operators into two parts

$$\psi(r,t) = \chi(r,t) + \tilde{\psi}(r,t), \tag{2.6}$$

$$\psi^{+}(r,t) = \chi^{+}(r,t) + \tilde{\psi}^{+}(r,t), \tag{2.7}$$

corresponding, one to the wave function of the condensate $\chi(r,t)$ and another for the non-condensed particles $\tilde{\psi}$. These could correspond, mainly, to thermal excitations and to quantum fluctuations. For a homogeneous system, $\chi(r,t)$ usually called as the condensate wave function which is a complex number including the order parameter ρ_0 and the phase ξ of the condensate:

$$\chi \equiv \xi\sqrt{\rho_0} \equiv e^{i\Theta}\sqrt{\rho_0}, \quad \chi^{+} \equiv \xi^{+}\sqrt{\rho_0} \equiv e^{-i\Theta}\sqrt{\rho_0}. \tag{2.8}$$

In an equilibrium system, the condensate wave function does not depend on time, $\chi(r,t) \equiv \chi(r)$.

The Grand thermodynamic potential of the system Ω with interacting triplons reaches its lowest value in the equilibrium. That is why we can minimize it in the below form

$$\frac{\partial\Omega}{\partial\rho_0} = 0, \quad \frac{\partial^2\Omega}{\partial\rho_0^2} > 0. \tag{2.9}$$

$$\frac{\partial\Omega}{\partial\theta} = 0, \quad \frac{\partial^2\Omega}{\partial\theta^2} > 0. \tag{2.10}$$

Fortunately, these equations do not contradict with the case $\langle\tilde{\psi}(r)\rangle = 0$, $\langle\tilde{\psi}^+(r)\rangle = 0$ [57; pp. 599-639. 58; pp. 062001- 41]. Here though it should be noted that, equations (2.10) impose a restriction to the phase of only the condensate wave function χ, while the phase of the $\tilde{\psi}$ in (2.7) , and therefore that of the wave function, corresponding to the operator ψ remains arbitrary in accordance with general laws of quantum mechanics. As stated above, our purpose is to calculate Ω, which includes almost all the information about the equilibrium statistical system, such as entropy $S = -(\partial\Omega/\partial T)$ and magnetization $M = -(\partial\Omega/\partial H) = g\mu_B N$ which can be found by this potential [59; pp.126313 – 8]. We write well-known formula for the grand thermodynamic potential

$$d\Omega = -SdT - PdV - Nd\mu - MdH. \tag{2.11}$$

Here, we use the path integral formalism where Ω can be found as

$$\Omega = -T\ln Z, \tag{2.12}$$

$$Z = \int D\tilde{\psi}^+ D\tilde{\psi} e^{-A[\psi,\psi^+]} \tag{2.13}$$

$$A[\psi,\psi^+] = \int_0^\beta d\tau d\vec{r} \left\{\psi^+\left[\frac{\partial}{\partial\tau} - \vec{K} - \mu\right]\psi + \frac{U}{2}(\psi^+\psi)^2 + +\frac{\gamma}{2}(\psi\psi + \psi^+\psi^+) + i\gamma'(\psi - \psi^+)\right\}. \tag{2.14}$$

In this integral τ is a period with $\beta = 1/T$. This integral can be evaluated by using some approximations. Here we use the variational perturbation theory [60; P01003 –

29. 61; pp. 251-275], or δ-expansion method.

According to δ-expansion method, we add to the action the following term

$$A_\Sigma = (1-\delta)\int d\tau d\vec{r}\left[\Sigma_n\tilde{\psi}^+\tilde{\psi} + \frac{1}{2}\Sigma_{an}\left(\tilde{\psi}^+\tilde{\psi}^+ + \tilde{\psi}\tilde{\psi}\right)\right] \tag{2.15}$$

Where $\delta = 1$ at the end of the calculations. The variational parameters Σ_n and Σ_{an} may be interpreted as the normal and anomalous self-energies, respectively. They are presented as:

$$\Sigma_n = (\Pi_{11}(0,0) + \Pi_{22}(0,0))/2, \tag{2.16}$$

$$\Sigma_{an} = (\Pi_{11}(0,0) - \Pi_{22}(0,0))/2, \tag{2.17}$$

$$\Pi_{ab}(\omega_n,\vec{k}) = (G(\omega_n,\vec{k}))^{-1}_{ab} - (G^0(\omega_n,\vec{k}))^{-1}_{ab} \tag{2.18}$$

and the Green functions $G(\omega_n,\vec{k})$, $G^0(\omega_n,\vec{k})$ are given by

$$G_{ab}(\tau,\vec{r};\tau',\vec{r}') = \frac{1}{\beta}\sum_{n,k} e^{i\omega_n(\tau-\tau\prime)+i\vec{k}(\vec{r}-\vec{r}\prime)}G_{ab}(\omega_n,\vec{k}) \tag{2.19}$$

$(a,b=1,2)$, where $\omega_n = 2\pi nT$ is the n th bosonic Matsubara frequency,

$$\sum_{n,\vec{k}} \equiv \sum_{n=-\infty}^{n=\infty}\int d\vec{k}/(2\pi)^3$$

and

$$G_{ab}(\omega_n,\vec{k}) = \frac{1}{\omega_n^2+E_k^2}\begin{bmatrix}\epsilon_k + X_2 & \omega_n \\ -\omega_n & \epsilon_k + X_1\end{bmatrix}. \tag{2.20}$$

In equation (2.20) E_k corresponds to the dispersion of quasi-particles (Bogolons)

$$E_k = \sqrt{\epsilon_k + X_1}\sqrt{\epsilon_k + X_2}. \tag{2.21}$$

where the self-energies X_1 and X_2 are given by

$$X_1 = \Sigma_n + \Sigma_{an} - \mu \tag{2.22}$$

$$X_2 = \Sigma_n - \Sigma_{an} - \mu \tag{2.23}$$

It is required that these self energies should be positive to denay E_k to be negative and remove unstability effects. Hence, $X_1 \geq 0$, $X_2 \geq 0$ and thy can be found by minimization of grand thermodynamic potential Ω

$$\frac{\partial\Omega}{\partial X_1} = 0, \quad \frac{\partial\Omega}{\partial X_2} = 0. \tag{2.24}$$

After some analytical calculations, by considering up to quadratic order in γ'^2, we obtain the following expression for Ω

$$\Omega = \Omega_{ISO} + \Omega_{EA} + \Omega_{DM}. \tag{2.25}$$

$$\Omega_{ISO} = -\mu\rho_0 + \frac{U\rho_0^2}{2} + \frac{1}{2}\sum_k (E_k - \epsilon_k) + T\sum_k \ln(1 - e^{-\beta E_k})$$
$$+\frac{1}{2}(\beta_1 B + \beta_2 A) + \frac{U}{8}(3A^2 + 3B^2 + 2AB), \tag{2.26}$$

$$\Omega_{EA} = \frac{\gamma\rho_0}{2}(\xi^2 + \bar{\xi}^2) + \frac{\gamma}{2}(B - A), \tag{2.27}$$

$$\Omega_{DM} = -i\gamma'(\bar{\xi} - \xi)\sqrt{\rho_0} - \frac{\gamma'^2}{X_2}, \tag{2.28}$$

where

$$\beta_1 = -\mu - X_1 + \frac{U\rho_0}{2}(\xi^2 + \bar{\xi}^2 + 4) \tag{2.29}$$

$$\beta_2 = -\mu - X_2 - \frac{U\rho_0}{2}(\bar{\xi}^2 + \xi^2 - 4) \tag{2.30}$$

$$A = T\sum_{k,n} \frac{\epsilon_k + X_1}{\omega_n^2 + E_k^2} = \sum_k W_k \frac{\epsilon_k + X_1}{E_k} \tag{2.31}$$

$$B = T\sum_{k,n} \frac{\epsilon_k + X_2}{\omega_n^2 + E_k^2} = \sum_k W_k \frac{\epsilon_k + X_2}{E_k} \tag{2.32}$$

and $\xi = e^{i\Theta}$, $\bar{\xi} = e^{-i\Theta}$, $W_k = (1/2)\coth(\beta E_k/2) = 1/2 + f_B(E_k)$ .Normal and anomalous densities are calculated as $\rho_1 = \int \langle \tilde{\psi}^+(r)\tilde{\psi}(r)\rangle d\vec{r}$ and $\sigma = \int d\vec{r}\langle \tilde{\psi}(r)\tilde{\psi}(r)\rangle$ respectively. As a result, we obtain the following expressions

$$\rho_1 = \frac{A+B}{2} = \sum_k \left[\frac{W_k(\epsilon_k + X_1/2 + X_2/2)}{E_k} - \frac{1}{2}\right] \equiv \sum_k \rho_{1k} \tag{2.33}$$

$$\sigma = \frac{B-A}{2} = \frac{(X_2 - X_1)}{2}\sum_k \frac{W_k}{E_k} \equiv \sum_k \sigma_k \tag{2.34}$$

And the total density and total magnetization can be found as

$$\rho = \frac{N}{V} = \rho_0 + \rho_1, \quad M = g\mu_B\rho. \tag{2.35}$$

By the condition (2.24) we obtain expressions for self energies

$$X_1 = 2U\rho + U\sigma - \mu + \frac{U\rho_0(\xi^2 + \bar{\xi}^2)}{2} + \gamma + \frac{2\gamma\prime^2 D_1}{X_2^2} \tag{2.36}$$

$$X_2 = 2U\rho - U\sigma - \mu - \frac{U\rho_0(\xi^2 + \bar{\xi}^2)}{2} - \gamma - \frac{2\gamma\prime^2 D_2}{X_2^2} \tag{2.37}$$

where

$$A_{1\prime} = \frac{\partial A}{\partial X_1} = \frac{1}{8}\sum_k \frac{(E_k W_{k\prime} + 4W_k)}{E_k} \tag{2.38}$$

$$A_{2\prime} = \frac{\partial A}{\partial X_2} = \frac{1}{8}\sum_k \frac{(\epsilon_k + X_1)^2(E_k W_{k\prime} - 4W_k)}{E_k^3} \tag{2.39}$$

$$B_{1\prime} = \frac{\partial B}{\partial X_1} = \frac{1}{8}\sum_k \frac{(\epsilon_k + X_2)^2(E_k W_{k\prime} - 4W_k)}{E_k^3} \tag{2.40}$$

$$D_1 = \frac{A_{1'}}{\bar{D}}; \quad D_2 = \frac{B_{1'}}{\bar{D}}; \quad \bar{D} = A_{1'}^2 - A_{2'}B_{1'} \tag{2.41}$$

$$W_{k'} = \beta(1 - 4W_k^2) = \frac{-\beta}{\sinh^2(\beta E_k/2)}. \tag{2.42}$$

For the Green's functions and self energies, we get the following expressions

$$G^{-1}(\omega_n, \vec{k}) = \begin{bmatrix} \epsilon_k + X_1 & -\omega_n \\ \omega_n & \epsilon_k + X_2 \end{bmatrix}, \tag{2.43}$$

$$(G^0(\omega_n, \vec{k}))^{-1} = \begin{bmatrix} \epsilon_k - \mu & -\omega_n \\ \omega_n & \epsilon_k - \mu \end{bmatrix}. \tag{2.44}$$

$$\Sigma_n = \mu + \frac{X_1 + X_2}{2}, \quad \Sigma_{an} = \frac{X_1 - X_2}{2}. \tag{2.45}$$

Now, Σ_n and Σ_{an} should satisfy Hugenholtz-Pines (HP) theorem without anisotropy in terms of X_1, X_2 as

$$\Sigma_n - \Sigma_{an} - \mu = X_2 = 0, \tag{2.46}$$

which reveals the Goldstone mode with a gapless energy dispersion

$$E_k = \sqrt{\epsilon_k + X_1}\sqrt{\epsilon_k + X_2}|_{X_2=0} = \sqrt{\epsilon_k(\epsilon_k + X_1)} \approx ck + O(k^3), \tag{2.47}$$

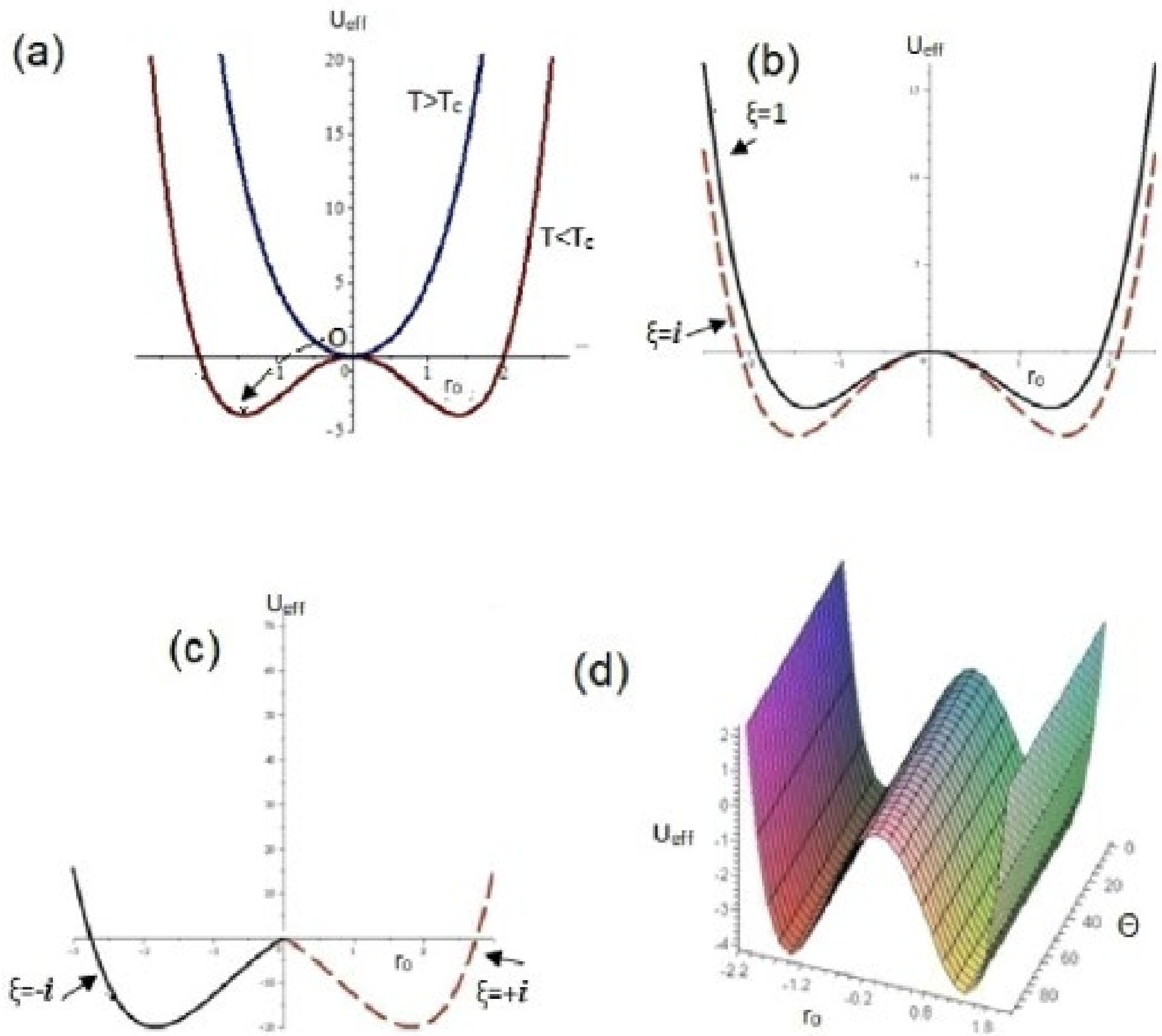
At this point, we want to discuss effective interaction potential at absolute zero temperature by neglecting quantum fluctuations and it takes the below form

$$U_{eff} = -\mu\chi\bar{\chi} + \frac{U\chi^2\bar{\chi}^2}{2} + \frac{\gamma(\chi^2 + \bar{\chi}^2)}{2} - i\gamma'(\bar{\chi} - \chi) \tag{2.48}$$

It is better to use unitless variables to azalyse this potential as

$$\widetilde{U}_{eff} = r_0^4 + 2r_0^2\left(\tilde{\gamma}\xi^2 + \tilde{\gamma}\bar{\xi}^2 - 2\right) + \frac{4ir_0\tilde{\gamma}\prime(\xi-\bar{\xi})}{\sqrt{\rho_{0c}}}, \tag{2.49}$$

where $\widetilde{U}_{eff} = 8U_{eff}U/\mu^2$, $r_0 = \sqrt{\rho_0/\rho_{0c}}$, $\tilde{\gamma} = \gamma/\mu$, $\tilde{\gamma}' = \gamma'/\mu$ and $\rho_{0c} = \mu/2U$ is the critical density of pure BEC. The result of isotropic case is given in Fig. 2.1 (a). In this case, while phase transition particle goes to the minimum point of the potential either to the left or right of the hole.



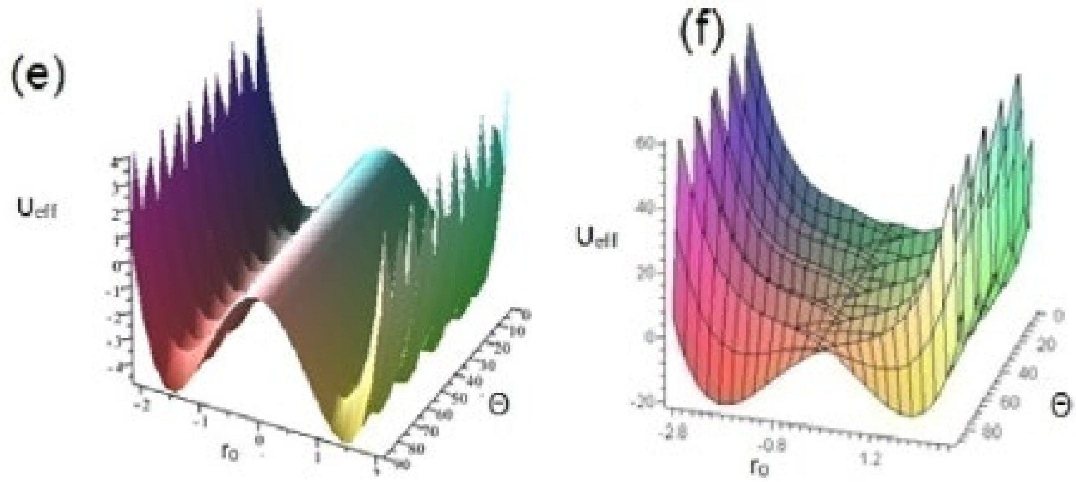


Fig. 2.1. $2D$ and $3D$ effective potentials for various cases of interaction: (a, d) $\gamma = \gamma' = 0$; (b, e) $\gamma \neq 0, \gamma' = 0$; and (c, f) $\gamma = 0, \gamma' \neq 0$

§ 2.3. The condensate phase and its density

The condensate fraction ρ_0 and phase Θ can be found by the equations (2.9-2.10) and (2.26-2.28) as

$$\frac{\partial \Omega}{\partial \rho_0} = \cos 2\Theta (U\sigma + \gamma) + U(\rho_0 + 2\rho_1) - \mu - \frac{\gamma' \sin\Theta}{\sqrt{\rho_0}} = 0, \tag{2.50}$$

$$\frac{\partial \Omega}{\partial \Theta} = 2\cos\Theta\left(2\rho_0 (U\sigma + \gamma)\sin\Theta + \gamma' \sqrt{\rho_0}\right) = 0, \tag{2.51}$$

$$-2\rho_0 \cos 2\Theta (U\sigma + \gamma) + \sin\Theta \gamma' \sqrt{\rho_0} > 0. \tag{2.52}$$

Here, the first equation is good to find condensate density while the other two are helpful to analyze phase angle. It can be seen that equation (2.51) has two solutions with respect to Θ which we refer these solutions as mode-1 and mode-2:

$$\Theta = \begin{cases} -\arcsin(\tilde{s}) + 2\pi n, & \xi = \sqrt{1-\tilde{s}^2} - i\tilde{s}: mode - 1 \\ \frac{\pi}{2} + \pi n, & \xi = \pm i: mode - 2 \end{cases} \tag{2.53}$$

where $\tilde{s} = \gamma'/2\sqrt{\rho_0}(U\sigma + \gamma)$, and $n = 0, \pm 1, \pm 2 \ldots$ may be interpreted as a topological number. For a while, we do not know which solution is physical. Moreover, we are required to discuss both modes according to the physical propertis of variables in these solutions. The interesting case occurs in mode-1 since in this case phase angle of the condensate wave function depends on some physical quantities such as $\gamma', \rho_0, U, \sigma$ and γ. This means that phase angle may change due to these parameters and physical observables if one proves this solution as acceptable case. We rewrite equations (2.36-2.37) and (2.50-2.52) in the following equivalent form:

$$X_1 = U\sigma + U\rho_0 + \gamma + \frac{(\xi^2+\bar{\xi}^2)(U\rho_0-\gamma-U\sigma)}{2} - \frac{i\gamma\prime(\xi-\bar{\xi})}{2\sqrt{\rho_0}} + \frac{2\gamma\prime^2 D_1}{X_2^2} \tag{2.54}$$

$$X_2 = -U\sigma + U\rho_0 - \gamma - \frac{(\xi^2+\bar{\xi}^2)(U\rho_0+\gamma+U\sigma)}{2} - \frac{i\gamma\prime(\xi-\bar{\xi})}{2\sqrt{\rho_0}} - \frac{2\gamma\prime^2 D_2}{X_2^2} \tag{2.55}$$

$$\mu = U\rho_0 + 2U\rho_1 + \frac{(\xi^2+\bar{\xi}^2)(U\sigma+\gamma)}{2} + \frac{i\gamma\prime(\xi-\bar{\xi})}{2\sqrt{\rho_0}} \tag{2.56}$$

$$\frac{\partial\Omega}{\partial\Theta} = (U\sigma + \gamma)(\xi^2 - \bar{\xi}^2)\sqrt{\rho_0} + i\gamma'(\xi + \bar{\xi}) = 0 \tag{2.57}$$

$$\frac{\partial^2\Omega}{\partial\Theta^2} = -2\rho_0(\xi^2 + \bar{\xi}^2)(U\sigma + \gamma) - i(\xi - \bar{\xi})\gamma'\sqrt{\rho_0} > 0 \tag{2.58}$$

Now we are on the stage to discuss both modes and find limits and restrictions for phase angle for various Bose gases. For example, we can apply these equations for ideal bose gas and in this case (2.57) become zero as we neglect interactions and anisotropy. Hence, phase of the BEC of an ideal Bose gas can take any value without any restriction and there is no any limit for this angle.

§ 2.3.1. Isotropic case: $\boldsymbol{\gamma = \gamma' = 0, U \neq 0}$

In this case solutions (2.53) will have two possible values for $\xi = \pm i$ and $\xi = 1$. And equation (2.57) has the form $U\sigma(\xi^2 - \bar{\xi}^2) = 0$. Hence, HFB approximation puts some restrictions for the phase while HFB approximation claims that it can take any value. Here, effective potential (2.48) does not depend on phase and satisfies the SSB case (see Figs. 2.1a and 2.1d). However, $\xi = \pm i$ with $\Theta = \pi/2$ case leads to $X_1 \leq 0$ and hence, to a maximum of thermodynamic potential where instability can occcur. That is why we should discuss mode-1 as a possible solution. In this case, with $\xi = 1,\ \Theta = 0,\ X_2 = 0$, our main equations take the following form

$$X_1 = 2U(\rho_0 + \sigma) \tag{2.59}$$

$$\mu = 2U\rho_1 + U\rho_0 - U\sigma \tag{2.60}$$

These equations give an acceptable solution [62; pp. 144416 – 12] as density of condensed quasiparticles ρ_0 have bigger value than anomalous density σ. This implies that phase angle of any BEC of interacting gases deserves values as $\Theta = \pi n$ [63; pp. 190404 -4. 64; pp. 167-170. 65; pp. 050603 -23]. Here, one should state that the sign of anomalous density is negative [66; pp.1-87. 67; pp. 043633–8. 68; pp. 033626–9].

§ 2.3.2. Quantum magnets with EA interaction

In present subsection, we analyze the case with $\gamma' = 0, \gamma \neq 0$. Firstly, we see the shape of the effective potential according to the different phases of condensed fraction whether it is real or complex (see Figs. 2.1 b and 2.1e).

$$U_{eff} = \begin{cases} -\mu\rho_0 + \frac{U\rho_0^2}{2} - \gamma\rho_0, & \xi = i \\ -\mu\rho_0 + \frac{U\rho_0^2}{2} + \gamma\rho_0, & \xi = 1. \end{cases} \tag{2.61}$$

The system reaches stability at the minimum of this potential and this lowest point can be found as $\rho_0 = (\mu \pm \gamma)/U$. So, the form of potential for this point with different ξ according to mode-1 or mode-2

$$U_{eff}^{min} = -\frac{(\mu-\gamma)^2}{2U}, \quad \xi = 1 \tag{2.62}$$

$$U_{eff}^{min} = -\frac{(\mu+\gamma)^2}{2U}, \quad \xi = i \tag{2.63}$$

Here, one can see a comparision of chemical potential μ and anisotropy parameter γ gives the results as $U_{eff}^{min}(\xi = i) < U_{eff}^{min}(\xi = 1)$ as illustrated in Figs. 2.1b and 2.1e. Moreover, equation (2.58) for the real phase will have the form

$$\rho_0(U\sigma + \gamma) < 0. \tag{2.64}$$

It means this equation is satisfied for any temperature for a small anisotropic parameter and for the case $U|\sigma| > \gamma$. This inequality also acceptable for low temperature. Hence, if we consider EA interactions only, the phase of the condensed fraction can take values as πn.

§ 2.3.3. EA and DM interactions: $\gamma' \neq 0, \gamma \neq 0$

Antiferromagnetic materials show some anisotropic properties, especially our compound $TlCuCl_3$ have a monoclinic crystal structure. Hence, we should consider both intra and inter dimer interactions. DM and EA interactions include these effects and we can study quantum magnets by these additional effects as a more realistic case.

The shape of effective potential is also plotted in Figs. 2.1c and 2.1f. Additionally, we can also analyze the phase angle of condensed triplons by these effects. From equation (2.53), it can be seen that phase angle has a dependence on anisotropic parameters and some other properties of the system as $\sin\Theta = -\gamma'/(2\sqrt{\rho_0}(U\sigma + \gamma))$. Interestingly, the phase angle may change due to temperature change as anomalous density formula includes theperature of the system. It means, there will be a dramatic change compared to the case with only EA. Since for the triplon system Θ corresponds to the angle between y-axis and staggered magnetization $M_\perp$ (see Fig. 2.3a), one may observe that changing the temperature leads to a change in σ and ρ_0, hence modifies the direction of $M_\perp$ in the (xy)-plane. In this case with the above phase angle, equations (2.54) and (2.55) give

$$X_1 = 2U\rho_0 - \frac{U\gamma'^2}{2(U\sigma+\gamma)^2} + \frac{2\gamma'^2 D_1}{X_2^2} \tag{2.65}$$

$$X_2 = -2(U\sigma + \gamma) + \frac{U\gamma'^2}{2(U\sigma+\gamma)^2} - \frac{2\gamma'^2 D_2}{X_2^2}. \tag{2.66}$$

It is easily understood that at high temperatures $(T \geq T_c)$ $X_1 \approx X_2$ and both are rather large $(X_2(T \gg T_c) \gg X_2(T = 0))$ [5]. Therefore, in this limit Eq. (2.65) leads to

$$X_1(T \gg T_c) \approx -\frac{U\gamma'^2}{2\gamma^2} \leq 0 \tag{2.67}$$

which corresponds to a dynamic instability. Moreover, Eq. (2.58) with Θ given by Eq. (2.53) is not satisfied, which means that, this solution corresponds to the maximum, not the minimum of Ω. We now discuss the case $\xi = -i$ and show that this should also be excluded. In fact setting $\xi = -i$ in (2.54) and (2.55) we obtain

[5] At high temperatures, the anomalous density given by Eq. (2.79) should vanish, and the energy $E_k \approx (\epsilon_k + X_2)$ is naturally expected to increase.

$$X_1 = 2U\sigma - \frac{\gamma\prime}{\sqrt{\rho_0}} + \frac{2\gamma\prime^2 D_1}{X_2^2} + 2\gamma \quad (2.68)$$

$$X_2 = 2U\rho_0 - \frac{\gamma\prime}{\sqrt{\rho_0}} - \frac{2\gamma\prime^2 D_2}{X_2^2}. \quad (2.69)$$

It is easily understood that, at high temperatures ($\rho_0 \to 0$), the second term in Eqs. (2.68) and (2.69) will dominate and hence, both of X_1 and X_2 become negative. Thus, we come to the conclusion that when $\gamma' \neq 0$ the only phase with $\xi = +i$ is accessible even in the HFP approximation.

Table 1

Possible phases and transitions in homogeneous BECs. SBS and EBS correspond to spontaneous and explicit breaking symmetry cases, respectively

BEC type	Interaction parameter	Phase	Phase angle	Symmetry breaking	Transition BEC → normal phase
Ideal gas	$U = 0, \gamma = 0, \gamma' = 0$	$\xi = e^{i\Theta}$	arbitrary	SBS	II-order
Pure BEC	$U \neq 0, \gamma = 0, \gamma' = 0$	$\xi = \pm 1$	πn	SBS	II-order
Interacting gas with EA	$U \neq 0, \gamma \neq 0, \gamma' = 0$	$\xi = \pm 1$	πn	EBS	II-order
Interacting gas with DM anisotropy	$U \neq 0, \gamma = 0, \gamma' \neq 0$	$\xi = +i$	$\pi/2 + 2\pi n$	EBS	crossover
Interacting gas with both EA and DM anisotropies	$U \neq 0, \gamma \neq 0, \gamma' \neq 0$	$\xi = +i$	$\pi/2 + 2\pi n$	EBS	crossover

The main equations Eqs. (2.54), (2.55) and (2.56) for the case $\gamma = 0$, $\gamma' \neq 0$ with $\xi = +i$ have the form

$$X_1 = 2U\sigma + \frac{\gamma\prime}{\sqrt{\rho_0}} + \frac{2\gamma\prime^2 D_1}{X_2^2} \tag{2.70}$$

$$X_2 = 2U\rho_0 + \frac{\gamma\prime}{\sqrt{\rho_0}} - \frac{2\gamma\prime^2 D_2}{X_2^2} \tag{2.71}$$

$$\mu = U(\rho_0 + 2\rho_1 - \sigma) - \mu - \frac{\gamma\prime}{\sqrt{\rho_0}}. \tag{2.72}$$

We shall discuss the properties of these equations in detail at our future works. Here we note that for $\gamma' \neq 0$ Eq. (2.72) has no solution with $\rho_0 = 0$, which prevents the existence of a critical temperature defined as $\rho_0(T = T_c) = 0$. Hence, there is no an ordinary phase transition from BEC to a normal phase but a crossover.

The results of this section are summarized in Table I. Thus we come to the conclusion that, similarly to the spontaneous magnetization phenomena, where at $T < T_{curie}$ the spins align in one direction without application of external magnetic field, a Bose gas at $T < T_c$ acquires a condensate fraction with a certain order parameter, ρ_0 and a certain phase, $\xi = e^{i\Theta}$ allowed by the conditions of stability against quantum and thermal fluctuations as illustrated in Fig. 2.1 (a).

§ 2.4. Interference of two condensates and Josephson effect

The best way of studying the phase of matter experimentally is through measurements with interference patterns or Josephson junctions. The former is usually performed with atomic Bose condensates and the latter with superconductors or, possibly, quantum magnets. Below we discuss the consequences of our results summarized in the previous section, to each of these effects.

§ 2.4.1. Interference between two Bose condensates

It is well known that the interference effect occurs due to the phase difference of two matter waves. Nearly ten years before observing the first interference effect between two BECs by the MIT group [54; pp. 637-641], Anderson [69; pp. 230-265] had raised his famous question “Do superfluids that have never seen each other have a well-defined relative phase?" Further this nontrivial question has been reformulated in a philosophical way “Does the BEC phase appears under the effect of SSB when it is formed, or later, when quantum measurement occurs?" There is no unique answer to these questions [70; pp. 880-887]. In fact, even if the phase preexists, it is doubtful wether it can be directly measured because of collisions during a ballistic expansion. Nevertheless, the existing experiments [71; pp. 120407 – 4. 72; pp. 060402 – 4. 73; pp. 1581-1584] seem to support that as a result of SSB the order parameter exists with a fixed phase, not only in homogenous infinite volume system, but also in inhomogenous system of finite size. This conclusion is consistent with our predictions derived in the previous section.

Qualitatively interference picture can be described in the following simple way. Suppose that we have an order parameter that involves two condensate wave functions, with condensate densities ρ_0^a and ρ_0^b in momentum states $\vec{k}_a$ and $\vec{k}_b$. Then, the density of the combined system is

$$\rho_0(\vec{r}) = |\sqrt{\rho_0^a} e^{i\vec{k}_a\cdot\vec{r}} e^{i\Theta_a} + \sqrt{\rho_0^b} e^{i\vec{k}_b\cdot\vec{r}} e^{i\Theta_b}|^2 = \rho_0^{ab}[1 + x\cos(\vec{k}\cdot\vec{r} + \Theta_{ab})] \quad (2.73)$$

Where $\rho_0^{ab} = \rho_0^a + \rho_0^b$, $\vec{k} = \vec{k}_a - \vec{k}_b$, $\Theta_{ab} = \Theta_b - \Theta_a$, $x = 2\sqrt{\rho_0^a \rho_0^b}/(\rho_0^a + \rho_0^b)$ and Θ_a, Θ_b are initial phase angels. So, we have an interference pattern with relative phase $\phi = \vec{k}\cdot\vec{r} + (\Theta_b - \Theta_a)$.

In Interference of two pure condensates case, as it is seen from Table I $\Theta_b - \Theta_a = \pi(n_b - n_a) \equiv \pi m$, $m = 0, \pm1, \pm2, \ldots.$. From Eq. (2.73) one obtains $\rho_0(\vec{r}) = \rho_0^{ab}(1 \pm \cos\vec{k}\vec{r})$. In particular, when both condensates have the same phase $\xi_a = \xi_b =$

$+1$, one obtains the well-known result, e.g., $|\vec{k}\vec{r}| = 2\pi n$ corresponds to constructive interference, since $\cos(\pi/2 - \pi n) = \sin\pi n$.

Now we consider BEC with DM interaction and suppose that we have two interfering condensates. One (b) is a pure condensate with SSB and the other (a) includes a tiny DM interaction, given by the linear Hamiltonian (2.5). The initial phases may be as $\Theta_a = \pi/2$ and $\Theta_b = 0$, so $\Theta_{ba} = -\pi/2$. Since $\cos(x - \pi/2) = \sin x$, (2.73) gives

$$\rho_0(\vec{r}) = \rho_0^{ab}(1 + x\sin(\vec{k}\vec{r})). \tag{2.74}$$

This means that, in contrast to the previous case, $|\vec{k}\vec{r}| = 2\pi n$ will correspond not to a constructive, but to a destructive interference. Thus, the presence of DM interaction in one of condensates dramatically changes the interference picture, demonstrating its sensitivity to the initial phase.

§ 2.4.2. Stationary Josephson effect

It is well known that Josephson effect can take place due to the phase difference $\Delta\Theta = \Theta_1 - \Theta_2$ between two contacting materials (d.c. effect) or due to the difference of chemical potentials $\Delta\mu = \mu_1 - \mu_2$ [74; pp. 10-48], (a.c. effect). Besides of superconductors, this effect has been observed in superfluid helium [75; pp. 280-283], where $\Delta\mu \neq 0$ is reached by application of a pressure differential, as well as in Bose condensates in a double well potential, by changing the relative condensate population $\Delta N = (N_1 - N_2)/N \sim \Delta\mu$ [76; pp. 579-583]. A natural question arises if Josephson effect is possible between two magnetic insulators, when each of them contains a triplon condensate. This question has been discussed some years ago by Schilling and Grundmann [41; pp. 2301-2309]. They predicted possible occurrence of Josephson effect between two compounds due to the chemical potential difference $\Delta\mu = g\mu_B(H_{c_1} - H_{c_2})$, say $Ba_3Cr_2O_8$ and $Sr_3Cr_2O_8$ separated by nonmagnetic $Ba_3V_2O_8$. However, the presence of a DM interaction has not been considered. Here we address the effect of DM interaction on the Josephson effect. First, we note that in the present article we have been discussing only equilibrium systems. Therefore, studying the a.c. Josephson effect, which is a dynamical effect, is beyond our scope. However, we may consider the d.c. Josephson effect. In the simple case with identical systems ($\Delta\mu = 0$) the change in number of triplons due to tunneling is given by

$$\frac{\partial \rho_{01}}{\partial t} = 2K\sqrt{\rho_{01}\rho_{02}}\sin(\Delta\Theta) \tag{2.75}$$

i.e., the current is simply proportional to the $\sin(\Delta\Theta)$ [77; pp. 325-340]

$$J_{dc} \approx \sin(\Theta_1 - \Theta_2). \tag{2.76}$$

Now, from Table I one may come to the conclusion that stationary Josephson effect can take place only when one of the materials has no anisotropy, (or only EA), $\Theta_1 = \pi n$ while the other one has DM anisotropy, $\Theta_2 = \pi/2 + \pi n$. In all other cases, e.g., in the contact between two materials with no anisotropy: $\Delta\Theta \approx \pi n$ (n=0, 1, 2...), and hence $J_{dc} = \sin(\pi n) = 0$. We hope, a proper choice of compounds with suitable material parameters will make the observation of Josephson effect possible in various regimes and in particular verify the above conclusion.

§ 2.5. Direction of transverse magnetization

One of the main effects on quantum magnets is its increased magnetization as temperature decreases below the critical temperature. It is said that this is the effect of BEC of triplons and the occurance of additional staggered magnetization which is perpendicular to the external magnetic field [78; pp.1387-1398 . 79; pp. 177-221. 80; pp.75-114. 81; pp. 117-150]. Additionally, phase of the condensed particles can explain this type of magnetization [82; pp. 948-951. 83; pp. 235-304. 84; pp. 2590 – 7. 85; pp. 312-315] as it is pointed out in the Introduction. The emergence of a triplon condensate in quantum magnets leads to a finite staggered magnetization $M_{\perp}$, whose magnitude may be evaluated as $|M_{\perp}| = g\mu_B\sqrt{\rho_0/2}$, where ρ_0 is the condensate fraction. As to the direction of the vector $\vec{M}_{\perp}$, it lies in the xy-plane as illustrated in Fig. 2.3a, so $M_{\perp} = |M_x|\sin\Theta + |M_y|\cos\Theta$. Remarkably, the angle Θ in this plane corresponds to the phase angle of the condensate wave function [86; pp.11398-11407].

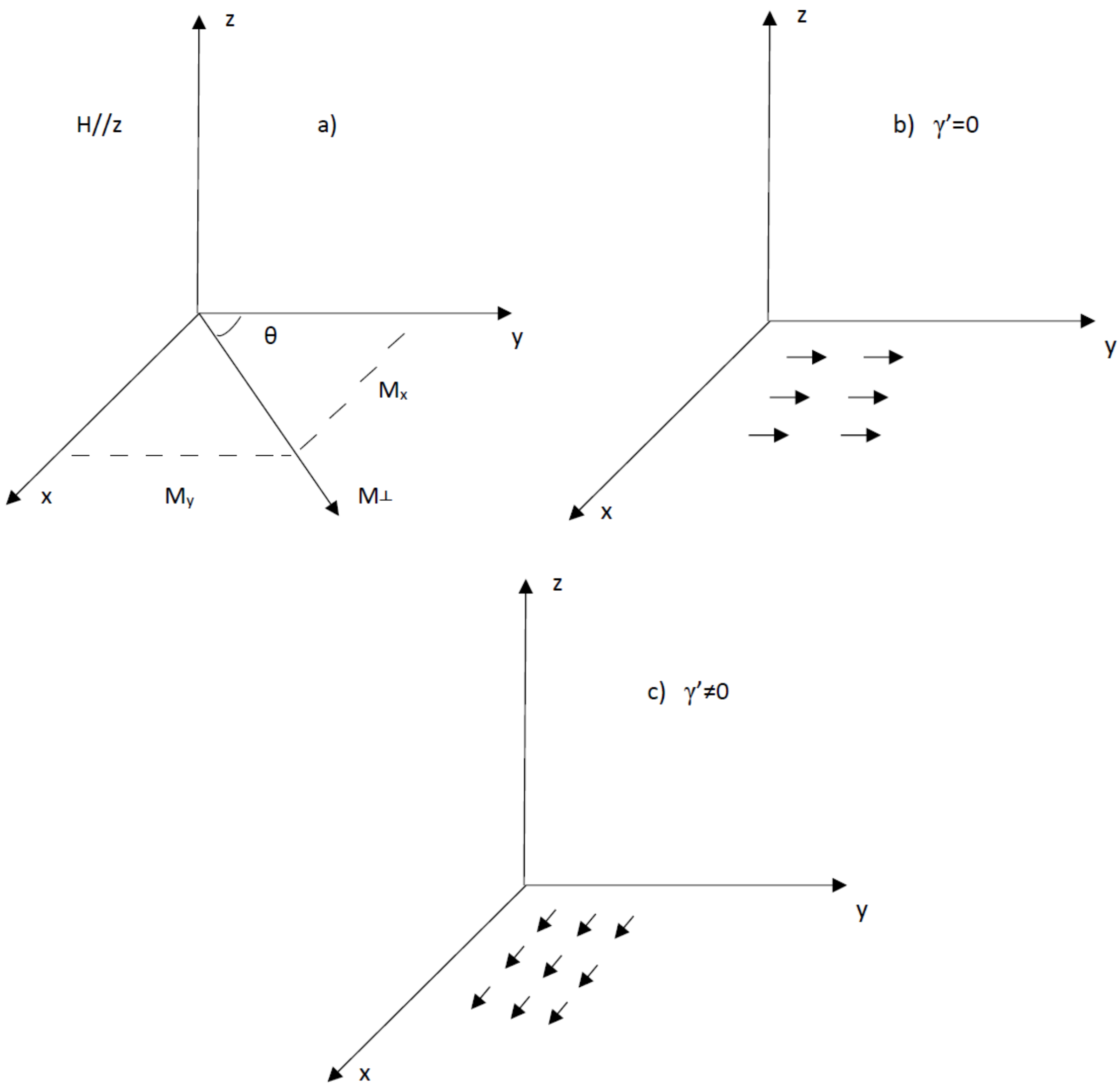


Fig. 2.2. The vector of staggered magnetization. (a) General representation; (b) and (c) correspond to the case of slow creation of triplon BEC without (b) and with DM anisotropy (c)

Thus, from Fig. 2.2b and Table I, one may conclude that $M_\perp$ lies along the y-axis ($\Theta = \pi n$) for the system without DM anisotropy, and $M_\perp$ is parallel to the x-axis, $\Theta = \pi/2$ when DM interaction is involved. It means that, in the presence of DM interaction each domain will have a phase angle $\pi/2 + 2\pi n$, $m_\perp = |m_x^i|$ which leads to the finite $M_\perp = \sum m_\perp^i = \sum |m_x^i| \neq 0$ (see Fig. 2.2c).

§ 2.6. Conclusion for Chapter II

We have derived an explicit expression for the grand canonical thermodynamic potential Ω for a triplon system in dimerized spin-gapped magnets, taking into account both the EA and weak DM anisotropies. The thermodynamic potential embodies all information about the equilibrium homogenous Bose gas at low temperatures. Satisfaction of these equations, together with stability conditions for BEC, present certain boundaries for the phase angle and condensate fraction of BEC within the framework of MFA. We have found that in the filed operator, ρ_0 has dependence on temperature and anisotropy but its phase is a constant value. But for the gases, this phase can have any angle and this is true even for the interacting gas also with $\sigma = 0$.

The system without anisotropy, or with only exchange anisotropy ($\gamma \neq 0, \gamma' = 0$) possesses only real phase with $\xi = \pm 1$, $\Theta = \pi n$, $n = 0,1,2...$ and BEC $\rightarrow$ normal phase transition takes place with a definite critical temperature T_c, such that $\rho_0(T \geq T_c) = 0$.

The inclusion of DM anisotropy smears this transition to a crossover, such that $\rho_0(T)$ diminishes asymptotically, and sets the phase to $\xi = +i$, rotating its angle Θ from 0 to $\pi/2$. There is a smooth path between BEC($\gamma \neq 0$)→ BEC($\gamma = 0$), in the sense that by gradually decreasing γ one arrives at a pure BEC. On the other hand, there is no path from a pure BEC to normal phase and crossover transitions: one may slowly decrease γ' to get the case with possible $\rho_0(T \geq T_c) = 0$, but the phase, which does not explicitly depend on γ' remains as $\xi = +i$, instead of $\xi = \pm 1$.

Therefore, our mean-field based approach, including the anomalous density predicts that the phase angle Θ of interacting homogenous BEC may take only discrete values as $\Theta = \pi n$ or $\Theta = \pi/2 + 2\pi n$, (n=0,$\pm$1, $\pm$2....) where n can be interpreted as a topological number. This is in contrast to widely used e.g., HFP or simple Bogoliubov approximations, where Θ is allowed to have any arbitrary angle.

We have shown that when one of the condensates have even a tiny DM interaction the interference picture will change drastically. Analyzing the simple d.c. Josephson effect between two spin-gapped magnets we have found that there would be no Josephson current when neither of samples has DM anisotropy $(\gamma'_1 = 0, \gamma'_2 = 0)$, while the current will be finite, when one of them has a weak DM anisotropy $(\gamma'_1 = 0, \gamma'_2 \neq 0)$.

We have shown that DM anisotropy destroys the total staggered magnetization in axially symmetric samples and in the presence of DM interaction $(\gamma' \neq 0)$, the staggered magnetization remains finite.

Presented results have been published in the following journals:

1. Rakhimov A., Khudoyberdiev A., Rani L., Tanatar B. Spin-gapped magnets with weak anisotropies I: Constraints on the phase of the condensate wave function //Annals of Physics – Elsevier (Netherland), 2021. – Vol. 424 – id.168361 -23 p.
2. Khudoyberdiev A., Narzikulov Z. The phase angle of the triplon condensate wave function with and without anisotropy // "Yadro fizikasi va Yadroviy texnalogiyalar", O’zbekiston yosh fiziklari VI respublika anjumani, 01-02 December– Tashkent (Uzbekistan), 2020 – pp. 59-65.
3. Abdurakhmonov T., Khudoyberdiev A. Restrictions on the phase angle of the triplon gas wave function // “Yosh olimlar va fizik talabalarning I Respublika ilmiy anjumani”, 21 April –Tashkent (Uzbekistan), 2021 –pp. 243.

III. Magnetization of antiferromagnetic materials with Dzyaloshinskii-Moriya and exchange anisotropies

§ 3.1 Introduction

We have seen in the previous chapters that some properties of quantum magnets can be well described by the BEC of quasiparticles. EA interactions explained the total magnetization of these materials near zero temperature and above the critical magnetic field. However, there were some artifacts of theoretical studies for experimental data on staggered magnetization. Because experimental results on antiferromagnetic materials showed an additional staggered (transverse) magnetization where it should have been zero at absolute zero temperature. This additional magnetization has become one of the interesting aspects of quantum magnets to describe theoretically [89; pp. 220402 – 4]. Neighbouring spins directions are antiparallel in antiferromagnetic materials. However, there might be some cantings and spins can be aligned with some angle to each other. In this case, there occur different types of interactions compared to exchange interactions. To include these antisymmetric interactions one has to consider Dzyaloshinskii-Moriya (DM) anisotropy of the crystal. Strong spin-orbit coupling and symmetry breaking causes antisymmetric interactions and helps to mix ground state and excited state of triplons. Also, one can use an external magnetic field to close the spectrum gap between excited and ground state. Theoretically, the excitation spectrum should be a gapless Goldstone mode associated with the spontaneous breaking of rotational symmetry by the staggered order and one can obtain $E_k \sim ck$, which is a convincing signal for the existence of BEC in this class of quantum magnets. For these reasons, low temperature thermodynamic effects of quantum magnets can be briefly described by the inclusion of DM interactions between canted spins of dimers [90; pp. 1145-1168].

As we are applying BEC theory for solid state, there should be some difference compared to BEC of gases. In atomic gases, total number of particles is considered as the constant number and it has the following form due to Bose statistic

$$N \sim \sum_k 1/[e^{(\varepsilon_k-\mu)/\mathrm{T}}-1] \tag{3.1}$$

By this formula, one tries to find chemical potential (μ). Hence, for atomic gases, the number of particles is the input parameter while the chemical potential of this system is output. On the other hand, for triplon system, it is vice-versa. The external magnetic plays role of chemical potential as well as it helps to minimize energy of S=+1 state and bond excited and ground states. It means, in the field induced BEC, μ is assumed to be an input parameter, from which the total number of triplons can be calculated. Hence, in triplon system, one needs to find the density of quasiparticles for the condensed and uncondensed part. Additionally, for atomic gases, kinetic energy of particles can be given by $\varepsilon_k = k^2/2m$. However, for quantum magnets more accurate form of dispersion is used (Appendix A) by considering the anisotropic properties of the system [91; pp. 565-571. 92; pp. 174406 – 6].

Usually, the total Hamiltonian of the system is written and diagonilized to find energy dispersion relation for low temperature theoretical analyzes. And it gives gapless Bogoliubov dispersion as $E_k = \sqrt{\varepsilon_k}\sqrt{\varepsilon_k + 2U\rho} \approx ck + O(k^3)$ with $\rho = N/V$ - density and c - sound velocity. But, low frequency electron spin resonance (ESR) measurements on some materials, such as $TlCuCl_3$ [93; pp. 020403 – 4. 94; pp. 184410 – 9], $(C_4H_{12}N_2)(Cu_2Cl_6)$ [95; pp. 054415-4], Cs_2CuCl_4 [96; pp. 037204-4], DTN [97; pp. 047205-4] gave results with a tiny spin gap. This evidence is the effect of anisotropy of the system such as exchange anisotropy (EA) or Dzyaloshinsky-Moriya (DM) interactions [15; pp. 275-281]. For this reason, we should include both types of anisotropic interactions for a brief theoretical description of low temperature properties of spin-gapped quantum magnets. As a result, with EA and DM interactions, our total Hamiltonian takes the following form

$$H_{aniz} = \int d\vec{r}\{\psi^+(r)(\hat{K} - \mu)\psi(r) + \frac{U}{2}(\psi^+(r)\psi(r))^2 + \frac{\gamma}{2}[\psi^+(r)\psi^+(r) + \psi(r)\psi(r)] + +i\gamma'[\psi(r) - \psi^+(r)]\} \tag{3.2}$$

where γ and γ' are parameters of EA and DM interactions, correspondingly ($\gamma \geq 0$, $\gamma' \geq 0$). In this chapter also, similar to previous one, we use HFB approximation by considering anomalous density $\sigma = \sum_k \sigma_k = \frac{1}{2}\sum_k \left(\langle a_k a_{-k}\rangle + \langle a_k^\dagger a_{-k}^\dagger\rangle\right)$. Additionally, to find other properties of triplon system such as magnetization, entropy and heat capacity we take grand thermodynamic potential (2.25) from Chapter II. In the previous chapter, it has been also analyzed that the field operator of condensed particles for triplons is complex [98; pp. 168361- 23. 99; pp. 593-600. 100; pp. 074712-13]. Hence, we have a ready grand thermodynamic potential for application and compare our theoretical results with experimental ones.

§ 3.2. Dispersion relation for spin-gapped quantum magnets

For dispersion relation of quasiparticles, self energies $X_{1,2}$ and the condensate fraction ρ_0 plays an important role. In bose gases, $X_2 = 0$ and Hugenholtz-Pines theorem is satisfied [101; pp. 23-32]. But for triplon system, both self energies should have positive values. The energy dispersion is given as $E_k = \sqrt{\varepsilon_k + X_1}\sqrt{\varepsilon_k + X_2}$. In this formula, self energies can be analyzed according to their physical meanings and acceptable estimations. Now, we rewrite the main equations of Chapter II:

$$X_1 = 2U\rho + U\sigma - \mu + \frac{U\rho_0(\xi^2+\bar{\xi}^2)}{2} + \gamma + \frac{2\gamma'^2 D_1}{X_2^2} \tag{3.3}$$

$$X_2 = 2U\rho - U\sigma - \mu - \frac{U\rho_0(\xi^2+\bar{\xi}^2)}{2} - \gamma - \frac{2\gamma'^2 D_2}{X_2^2} \tag{3.4}$$

$$\mu = U\rho_0 + 2U\rho_1 + \frac{(\xi^2+\bar{\xi}^2)(U\sigma+\gamma)}{2} + \frac{i\gamma'(\xi-\bar{\xi})}{2\sqrt{\rho_0}} \tag{3.5}$$

where

$$A_{1\prime} = \frac{\partial A}{\partial X_1} = \frac{1}{8}\sum_k \frac{(E_k W_{k\prime} + 4W_k)}{E_k} \tag{3.6}$$

$$A_{2\prime} = \frac{\partial A}{\partial X_2} = \frac{1}{8}\sum_k \frac{(\epsilon_k + X_1)^2 (E_k W_{k\prime} - 4W_k)}{E_k^3} \tag{3.7}$$

$$B_{1\prime} = \frac{\partial B}{\partial X_1} = \frac{1}{8}\sum_k \frac{(\epsilon_k + X_2)^2 (E_k W_{k\prime} - 4W_k)}{E_k^3} \tag{3.8}$$

$$D_1 = \frac{A_{1\prime}}{\overline{D}}; \quad D_2 = \frac{B_{1\prime}}{\overline{D}}; \quad \overline{D} = A_{1\prime}^2 - A_{2\prime} B_{1\prime} \tag{3.9}$$

$$W_k = \frac{\coth(\beta E_k/2)}{2}; \quad W_{k\prime} = \beta(1 - 4W_k^2) = \frac{-\beta}{\sinh^2(\beta E_k/2)}. \tag{3.10}$$

$A = \rho_1 - \sigma$, $B = \rho_1 + \sigma$, ρ_1 - the normal and σ - anomalous densities. The quantities $X_{1,2}$ are related to the ordinary normal, Σ_n, and anomalous, Σ_{an}, self energies as follows: $X_{1,2} = \Sigma_n \pm \Sigma_{an} - \mu$.

§ 3.3. Self energies and strength of DM interaction

From the above (3.3) and (3.4) equations, it can be seen that these self energies have a dependence on temperature, external magnetic field, phase of the condensate wave function and anisotropic parameters. Their value may change due to these effects. However, we have found fixed values of phase for triplon system as $\xi = i$ (Chapter II Table I). One of our main interests is to find characteristics of the temperature dependence of these energies. The main equations for self-energies X_1 and X_2 are obtained from (3.3) and (3.4) by setting $\xi = i$,

$$X_1 = 2U\sigma + 2\gamma + \frac{\gamma\prime}{\sqrt{\rho_0}} + \frac{2\gamma\prime^2 D_1}{X_2^2} \tag{3.11}$$

$$X_2 = 2U\rho_0 + \frac{\gamma\prime}{\sqrt{\rho_0}} - \frac{2\gamma\prime^2 D_2}{X_2^2}. \tag{3.12}$$

Here, we have one more undefined quantity ρ_0 and this condensate density can be found by (3.5). We will present in the following dimensionless compact form of this equation:

$$r_0^3 + Pr_0 + Q = 0 \tag{3.13}$$

where we have introduced $P = -\bar{\sigma} + 2(\bar{\rho}_1 - 1 - \bar{\gamma})$, $Q = -2\bar{\gamma}'/\sqrt{\rho_{c0}}$, $r_0^2 = \rho_0/\rho_{c0}$, $\bar{\sigma} = \sigma/\rho_{c0}$, $\bar{\rho}_1 = \rho_1/\rho_{c0}$, $\bar{\gamma} = \gamma/\mu$, $\bar{\gamma}' = \gamma'/\mu$ and ρ_{c0} is the critical density of pure BEC, $\rho_{c0} = \mu/2U$. In general, one has to solve these three coupled nonlinear algebraic equations with respect to X_1, X_2 and r_0 at a given temperature and magnetic field. Clearly, in such cases it is important to guess initial values of $X_1(T)$ and $X_2(T)$, since the solutions are not unique. We have solved these problems with the Fortran program and analyzed them below and above critical temperature for different approximantions such as HFB and HFP.

In Fig. 3.1 , we present typical solutions of Eqs. (3.11)-(3.13) versus temperature for $g = 2.06$, $U = 315$ K, $H = 8.5$ T, $\gamma' = 0.1$ K and $\gamma = 0$. It can be seen that the effect of the 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.3.1(c). Because consideration of DM interactions changes the shape of self energy temperature dependent graphs. Both X_1, X_2 have considerable values above the critical temperatures. However, X_2 is more higher valuse through all temperature regime compared to X_1, ($X_1/X_2 \approx 10^{-4}$). Interestingly, this comparison is not acceptable for HFP approximation where $\sigma = 0$.

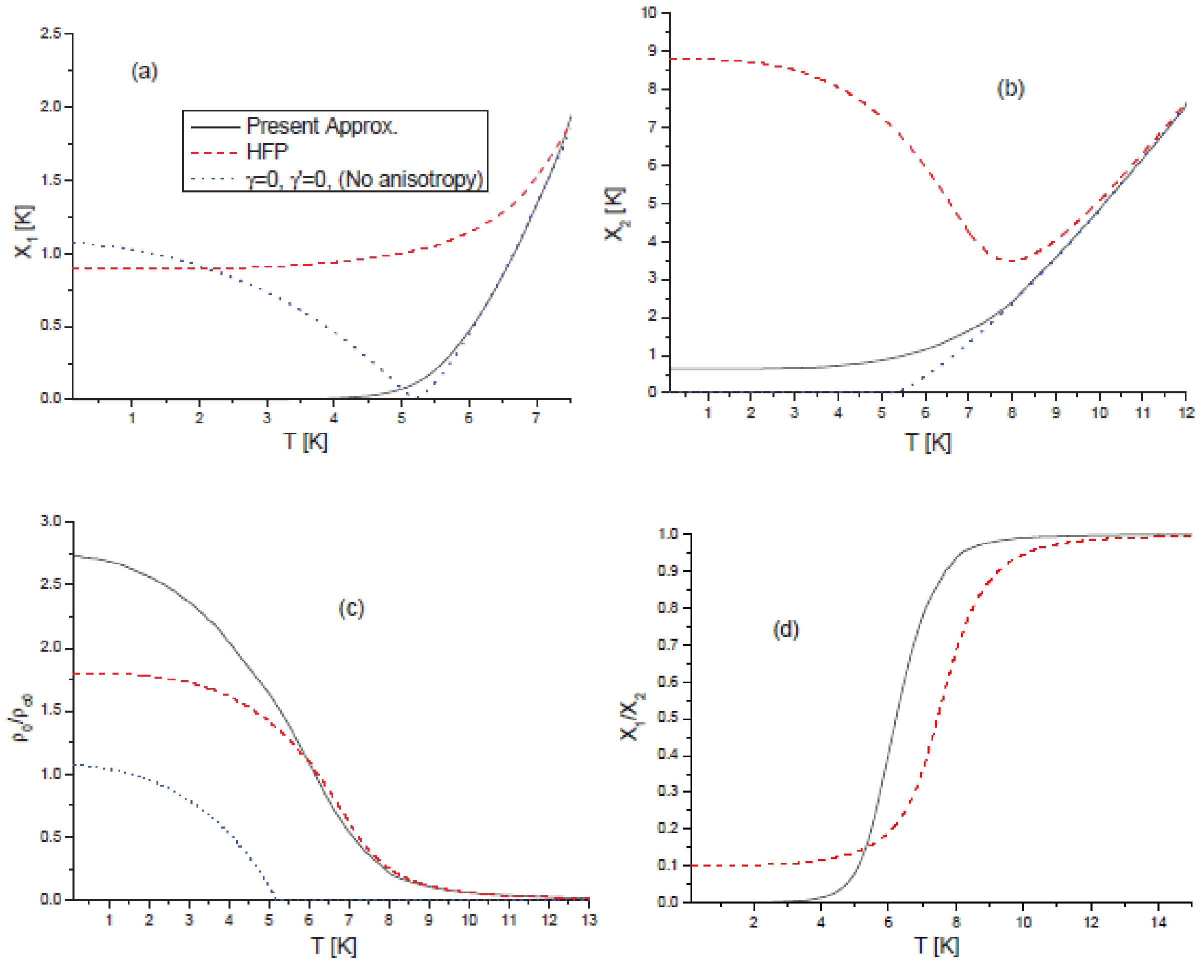


Fig.3.1. Figures (a), (b) and (c) show the self energies $X_1(T)$, $X_2(T)$ and condensate fraction $\rho_0(T)/\rho_{0c}$ ($\rho_{0c} = \mu/2U = 0.07$) dependence on temperature, respectively, while (d) illustrates the ratio $X_1(T)/X_2(T)$. Solid and dashed lines correspond to HFB and HFP approximation, correspondingly. The dotted lines represent the isotropic case with $\gamma = \gamma' = 0$

Below the critical temperature, transverse magnetization is present and this can be explained by condensate fraction. Additionally, our results show that condensate density never reaches zero even above the critical point. Hence, transverse magnetization remains finite in this region, also. In fact, since in the presence of DM interaction, $Q \neq 0$, Eq. (3.13) does not have a zero solution, as illustrated in Fig. 3.1(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.3.1(c) one may conclude that, DM anisotropy smears out BEC transition into a crossover.

From (3.11) it can be immediately seen that, since $\sigma > 0$, [6] $X_1(T \to 0) \neq 0$ when $\gamma' \neq 0$, that is the gap in the quasiparticle dispersion $E_k = \sqrt{(\varepsilon_k + X_1)(\varepsilon_k + X_2)}$ can never be closed for $\gamma' \neq 0$. 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 (3.11) with $D_2 > 0$, since lowering the temperature leads to decreasing also X_2. Here, it should be noted that we have taken into account DM interactions up to second order in (3.11) and (3.12) with γ'^2. This DM interaction parameter has dramatic effects on the equations for its large values with γ'^2. Now, we can state that DM interaction should be weak and DM parameter should be small enough. We have found some optimal values of this DM parameter for $TlCuCl_3$ with $\gamma'_{optimum} \approx 0.02$ K and its maximum is $\gamma'_{max} \approx 0.7$ K. Otherways, DM interactions destroy condensate state and BEC theory do not work properly. Moreover, one can get more effective and accurate results for extended orders of γ'.

§ 3.4. Properties of thermodynamic parameters

In the first chapter, we have considered only EA interactions and obtained some results for the physical quantities of the triplon system. In the second chapter, we have also included DM interaction and found that this antysmmetric anisotropy is important for the theoretical description of the qunatum magnets with BEC of triplons. DM interactions modify phase of the triplon condensate wave function operator which is totally different from BEC of gases. These interactions change second order phase transition to crossover at the critical temperature. Now, we aim to study some physical properties of quantum magnets with additional H_{EA} and H_{DM} Hamiltonians as in the (3.2). Moreover, we try to explain experimental data on staggered and total

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[6] See the next section

magnetization for the whole temperature regime ($T < T_c$, $T = T_c$, $T > T_c$) for antiferromagnetic compound *TlCuCl₃*.

§ 3.4.1. Anomalous fraction

Anomalous density is the result of pair-correlated interactions of triplons. There is only theoretical descriptions with formulas for this quantity as in equations (1.19) and (2.34). Experimentally, it has not been measured yet. In the first Chapter, we have obtained graphs of anomalous density dependence on temperature (Figs. 1.3) and stated that its value is negative for pure BEC. It is also compared to condensate fruction which is worth to consider for theoretical studies. In this subsection, we analyze the effect of DM interaction on this σ. In this case, we have the following formula

$$\sigma = U\sum_k \frac{W_k}{E_k}\frac{[\rho_0 - \gamma\prime^2 (D_1 + D_2)/UX_2^2]}{1 + U\frac{W_k}{E_k}} \tag{3.14}$$

From equation (3.9), one can see that $(D_1 + D_2) \leq 0$. All other terms are positive in (3.14) with energies, condensate fruction and DM interaction parameter. It means that $\sigma \geq 0$ at any temperature for $U > 0$, $\gamma' > 0$. Moreover, DM interactions totally modify anomalous density for HFB approximation.

The temperature dependence of σ calculated numerically by (3.14). The results for the ratio σ/ρ are given in Fig. 3.2 for parameters $g = 2.06$, $U = 315$ K, $H = 8.5$ T with similar graph as in Fig 1.3 (a). It can be seen that σ can be compared with total density of DM interacting triplons which is accounted for about 30 % of ρ for reliable γ'.

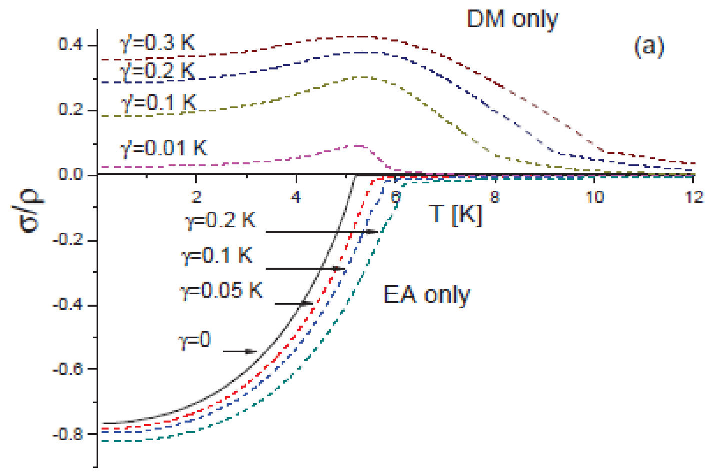

Fig.3.2. The ratio of anomaly density σ to the total density ρ of triplons vs temperature for various intensity parameters of DM and EA interactions. It is seen that presence of DM interaction changes the sign of anomalous density to opposite

§ 3.4.2. Magnetization

In this section, we present analytical and numerical results of magnetization with DM and EA interactions. We analyze their dependence on temperature above and below the critical one with the same input parameters as in Figs.3.1. We showed that there are two types of magnetization, total ($M = g\mu_B\rho$) and staggered ($M_\perp = g\mu_B\sqrt{\rho_0/2}$) one which can be found by total density and condensate density of triplons, respectively. It can be seen that these two magnetizations have sensitive dependence on EA interactions (γ) below the critical temperature where $T \leq T_c$ (Figs.3.3 (a), (c)). However, DM anisotropy has a significant effect on both total and staggered magnetization even above T_c. Additionally, in contrast to EA interaction, the strength of DM interaction (γ') prevents the staggered magnetization from vanishing at $T \geq T_c$ (Figs.3.3 (b), (d)). It means that we have good numerical results with some fitting parameters for staggered magnetization to describe experimental data announced by Tanaka *et al.* some years ago [37; pp. 939 -942]. Here, one has to know how to find the critical temperature for this result. Because in the first chapter, while considering only EA, we chose this point where condensate fraction becomes zero. But, in the case of DM interactions, the minimum point of total magnetization can be considered as the critical temperature for the occurrence of condensate. Our results for staggered magnetization confirm the conclusion of Chapter II as we stated that above the critical temperature there will be condensate fraction and there is a crossover instead of second order phase transition.

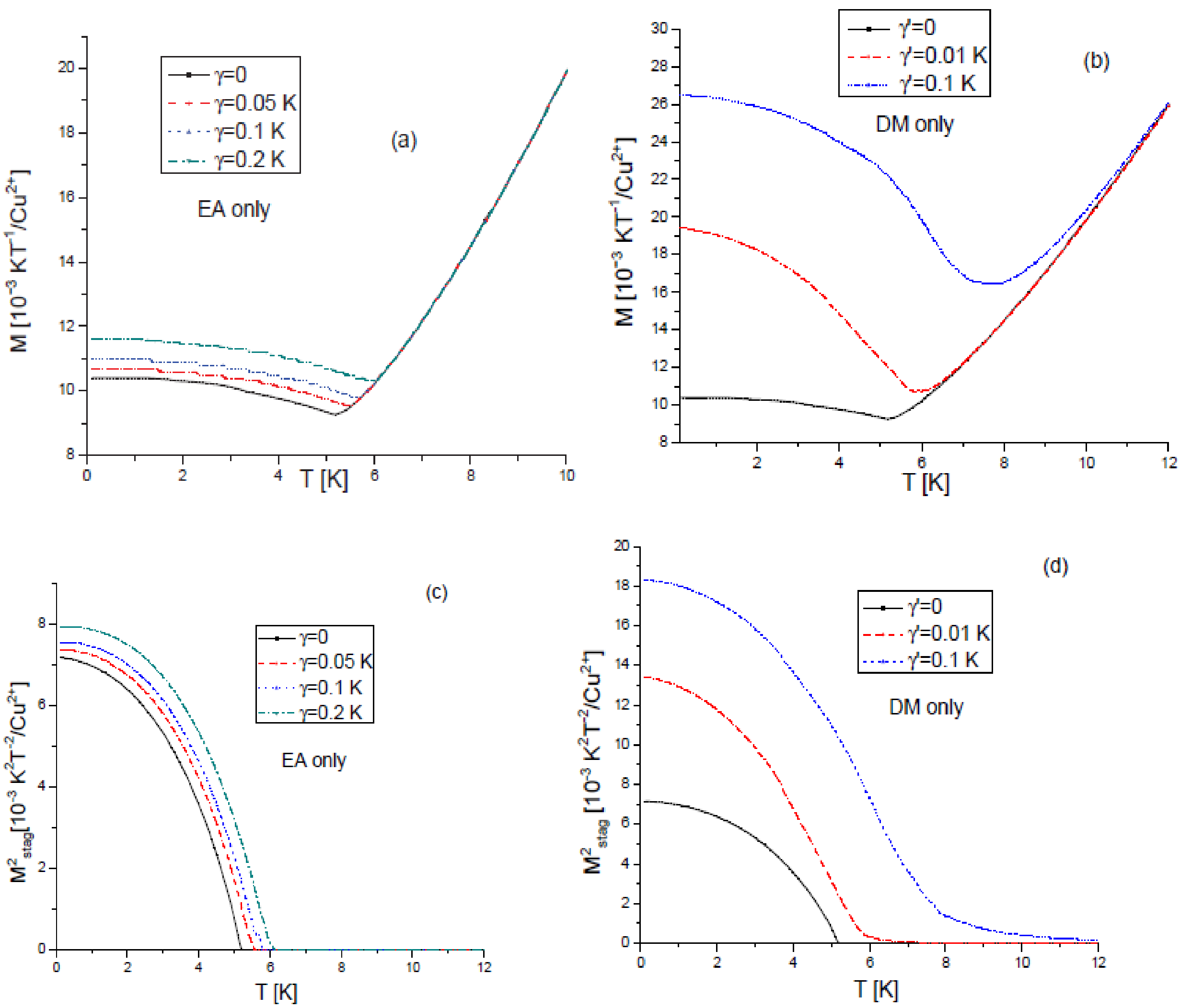

Fig.3.3. Total magnetization dependence on temperature with EA (a) and DM (b) interaction, separately. Solid lines represent the results for $\gamma = \gamma' = 0$. Figures (c) and (d) present the square of staggered magnetization $(M_{\perp})^2$

§ 3.4.3. Specific heat at the constant magnetic field, C_H

Analyzation of the temperature dependence of heat capacity is one of the best ways to study phase transition. Moreover, the phase transition from the normal state to BEC state can be well described by heat capacities of these two matter of state in the region of critical temperature. It has been found that the temperature dependence of this quantity has discontinuity ($\Delta C_V \equiv \lim_{\epsilon\to 0}[C_V(T_c-\epsilon)-C_V(T_c+\epsilon)] \neq 0$) and a well-known λ-shape [105; pp. 320-346] at T_c. Usually, specific heat is measured at constant volume and written as C_V. But, in this work, we have studied the heat capacity of triplons at constant magnetic field and marked as C_H. Our aim is to analyze this quantity in the presence of anisotropies. However, in the presence of DM interaction, calculation of C_H will have some difficulties as many quantities have a dependence on temperature in the grand thermodynamic potential (2.25). We will see its effects in our future works. Here, we discuss the results of the inclusion of only EA interactions.

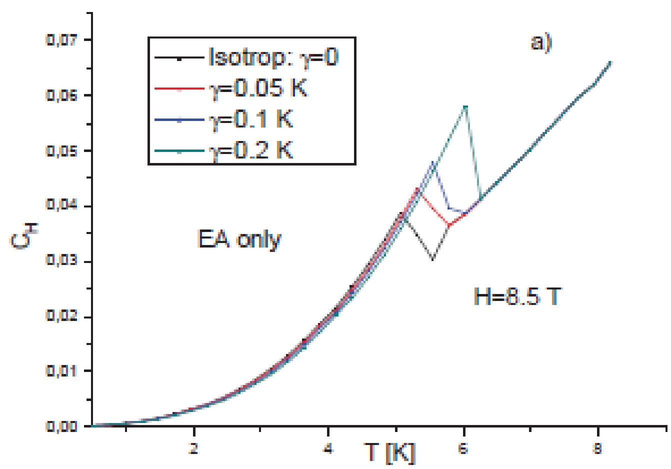

Fig.3.4. The heat capacity C_H dependence on temperature with only EA anisotropy. The input parameters are the same as in Figs. 3.1

Our obtained numerical results for $C_H(T)$ (detailed calculations are given at our article [106; pp. 2150223-23]) show that the isotropic and anisotropic cases give the same shape for specific heat except in the region on critical temperature (see Fig.3.4).

Hence, the anisotropic effects are important mainly in the critical region with its expected λ-shape view and there is a clear point for T_c where $\rho_0(T = T_c) = 0$, which separates BEC and normal phases. While EA leads to a sharp maximum in the specific heat (see Fig. 3.4(a)), in the case of a pure BEC without any anisotropy it also possesses fluctuations at the critical temperature as expected (solid black line in Fig. 3.4(a)). Additionally, anisotropy have significant effect not only on C_H but also on T_c. Critical temperature increases as the strength of anisotropy γ goes up.

§ 3.5. Results of anisotropic parameters for $TlCuCl_3$

In this section, we compare our results with experimental data. In the first Chapter I, we had good results for the description of total magnetization data. But, our results could not explain experimental results for staggered magnetization in the region of $T \geq T_c$. The reason for this is that we considered only EA interaction. However, in this chapter, we have also included DM interaction and we are expecting more accurate results for staggered magnetization. The anisotropic parameters γ and γ' can be fitted with realistic values for real antiferromagnetic materials.

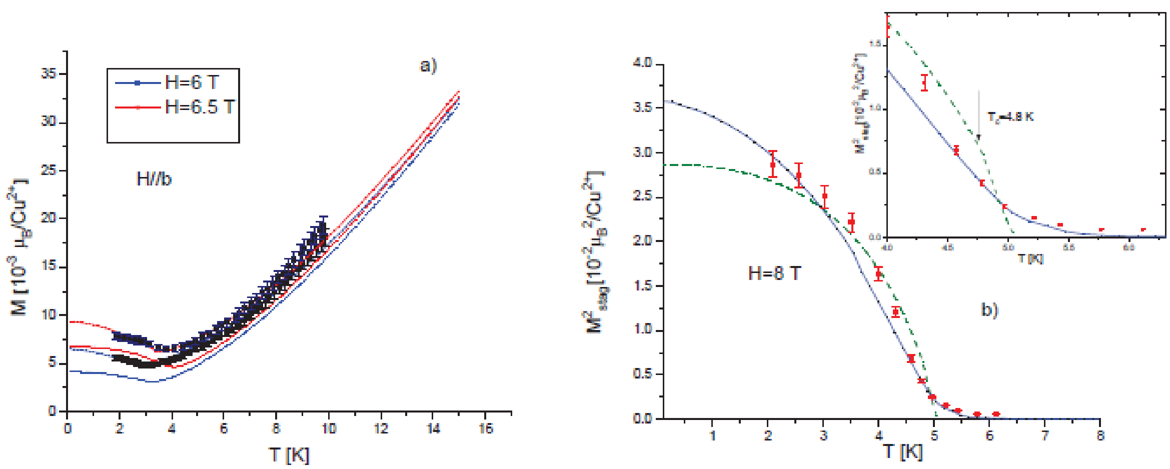

Fig. 3.5. Total (a) and staggered (b) magnetizations for $TlCuCl_3$. Solid and dashed lines correspond to HFB and HFP approximations, respectively. Experimental data are taken from [32; pp. 224403-8]. The optimized parameters are $\gamma = 0.05$ K, $\gamma' = 0.0201$ K and $U = 367$ K

Among the 3D quantum dimerized magnets with a spin gap $TlCuCl_3$ seems to be the most experimentally studied compound [107; pp.134416-4. 108; pp. 094426-7]. The observation of a finite $M_\perp$ at $T \geq T_c$ uniquely indicates the presence of DM interaction with a finite γ'. Thus, using existing experimental data on the magnetization and the heat capacity of $TlCuCl_3$, 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. 3.5 (a) and (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.3.5(b)). Moreover, taking into account the anomalous density σ leads to a better description of M e.g. at low temperatures, compared with the Sirker's approximation, where σ has been neglected.

To conclude, we have found that, the experimental data on magnetization and specific heat of $TlCuCl_3$ can be well described by the present approach.

§ 3.5.1. Energy dispersion for quasiparticles

There exist experimental measurements on the energy of magnetic excitations. Inelastic neutron scattering experiments help to study energy dispersion of triplons in some quantum magnets [94; pp. 184410 – 9]. Spin gapped quantum magnet e.g. $TlCuCl_3$ has a dimer structure and a finite energy gap in zero field Δ_{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. 3.6 (a).

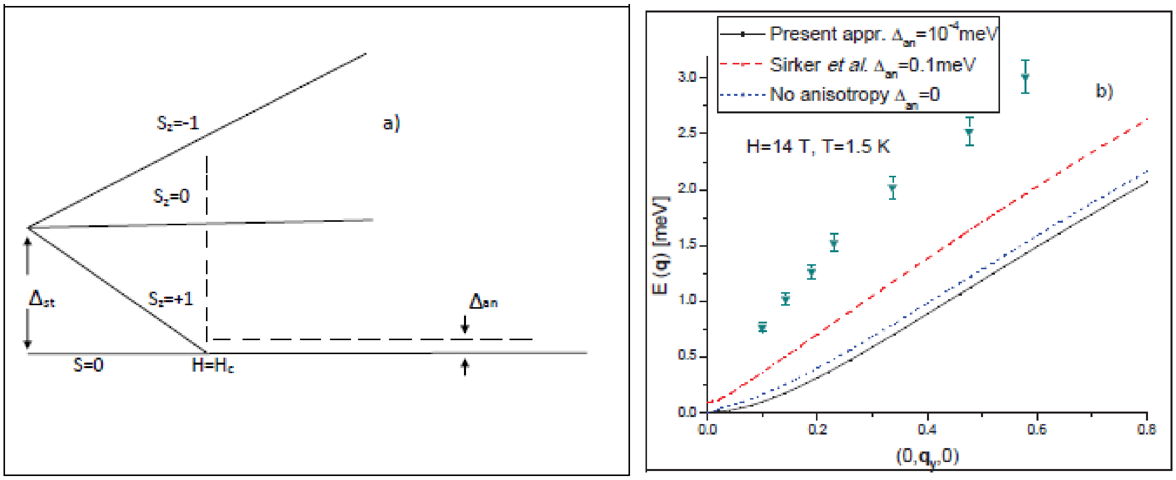

Fig. 3.6. a) The schematic illustration of energy levels of a spin-gapped system. At H=H $_c$ the gap Δ_{ST} closes and may reopen due to anisotropies with a tiny gap Δ_{an}; b) Energy dispersion of the low-lying magnetic excitations in $TlCuCl_3$. The solid, dashed and dotted lines correspond to present approximation including anisotropies, HFP approximation and without anisotropy , respectively. The experimental data are taken from [5; pp. 62-65]

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 as 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 $\Delta_{an} \sim 0.2$ meV, which was not observed in INS experiments (see Fig.3.6a). Therefore, the experimental situation on the energy spectrum of $TlCuCl_3$ has not been clear yet. In fact, on the one hand , the lowest excitation spectrum for $H_c \leq H \leq H_{saturation}$ at $T \leq T_c$ is gapless, $\Delta_{an}(INS) = 0$, on the other hand, it has a finite gap $\Delta_{an}(ESR) \neq 0$ and hence can not be linear. Theoretically, it is clear that, if the gap remains finite it may be caused by a lattice anisotropy.

The dispersion of elementary excitations at zero field ε_k is well studied experimentally [108; pp. 094426 -7] and presented as a function of momentum and intra (inter) - dimer interactions J_i as $\varepsilon_k(J_i)$. For theoretical studies, we have used more realistic dispersion of collective excitations which is given in Appendix A. One can find in the literature an explicit expression for $\varepsilon_k(J_i)$ with its optimized parameters.

As to the energy spectrum at $H \geq H_c$ it is clearly model dependent. For example, in the isotropic case at $T \leq T_c$ it is gapless, given by $E_k = \sqrt{\varepsilon_k + X_1}\sqrt{\varepsilon_k} \sim ck + O(k^3)$, so, $\Delta_{an} = E_k|_{k\to 0} = 0$ in agreement with experimental predictions by Rüegg et al. [5; pp. 62-65]. In the presence of anisotropies it has a finite gap $\Delta_{an} = \sqrt{X_1 X_2}$, where X_1, X_2 are defined by the system of equations (3.19) and (3.20). By using our optimal input parameters we obtained a finite but rather small value $\Delta_{an}(H = 14$ T, T=1.5 K)=10 $^{-4}$ meV, which is consistent with INS measurements, but not with ESR ones :$\Delta_{an}(H = 14$ T, T=1.5 K)=0.2 meV . In Fig. 3.7(b) we present quasiparticle spectrum $E_k = \sqrt{\varepsilon_k + X_1}\sqrt{\varepsilon_k + X_2}$, $(k_x = k_z = 0, k_y = \pi q_y)$ for $H = 14$ T at $T = 1.5$ K. It is seen that, the excitation energy in present approximation is almost linear, in accordance with experimental predictions. However, the values of $E_k^{exper.}$ are rather underestimated. This can be understood as follows. As it has been shown in Section 3.2 at low temperatures, the self energies, especially X_1 is rather small (Fig.1). Our input parameters optimized by experimental magnetisations lead to much smaller values: $X_1(H = 14$ T, T=1.5 K)=0.67·10 $^{-5}$ K, $X_2(H = 14$ T, T=1.5 K)=0.19 K, so $X_1 \ll X_2$. As a result, the momentum dependence of the dispersion is similar to that of isotropic one: $E_k = \sqrt{\varepsilon_k + X_1}\sqrt{\varepsilon_k + X_2} \sim \sqrt{\varepsilon_k}\sqrt{\varepsilon_k + X_2}$ 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 the presence of EA and DM interactions. Note that, a similar situation has been observed for compounds $Sr_3Cr_2O_8$ and $Ba_3Cr_2O_8$, who has DM interaction, but no anisotropy gap, i.e. $\Delta_{an}(Sr_3Cr_2O_8) = 0$, $\Delta_{an}(Ba_3Cr_2O_8) = 0$.

According to results of magnetization (Fig.3.5) and energy spectrum (Fig.3.6 (b)), it can be stated 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. Anyway, the main reason of our failure seems to be the simplicity of the Hamiltonian H_{DM} used here (the last term in Eq. (3.2)). 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 $H \perp (1,0,\bar{2})$ either.

Conclusion for Chapter III

We have studied effects of lattice anisotropies on thermodynamic characteristics of spin-gapped quantum magnets. 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 ρ_0. We have concluded 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 anisotropy strongly modifies the condensate fraction converting BEC transition into a crossover.

At low temperatures the DM interaction increases ρ_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 "λ-shape" of the heat capacity disappears due to strong DM interactions. On contrary, the presence of only EA anisotropy leaves the "λ-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 exchange anisotropy.

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

Presented results have been published in the following journals:

1. Rakhimov A., Khudoyberdiev A., Tanatar B.. Effects of exchange and weak Dzyaloshinsky-Moriya anisotropies on thermodynamic characteristics of spin-gapped magnets // International Journal of Modern Physics B – World Scientific(Singapore), 2021 –Vol. 35 –id.2150223-23 p.
2. Khudoyberdiev A., Rakhimov A. Anisotropic properties of antiferromagnetic materials at low temperatures // International Conference "Fundamental and applied problems of Physics", 22-23 September – Tashkent (Uzbekistan), 2020 – pp. 29.
3. Xudoyberdiev A. Triplonlarning dzyaloshinsky-moriya va almashinuv anizatropiyalari tasiridagi boze enshteyn kondensatsiyasi // "Yadro fizikasi va yadroviy texnologiyalar", O'zbekiston yosh fiziklari V respublika anjumani, 4-5 December –Tashkent (Uzbekistan), 2018–pp. 65-66.

CONCLUSION

We have studied low temperature phase transitions and magnetization at anisotropic quantum magnets. The obtained results can be summarized as follows:

1. We proposed a new mean-field approximation based approach within the Hartree-Fock-Bogolyubov approximation, which takes into account an anomalous density σ and anisotropy of the system. The low temperature properties of quantum magnets can be well described within this approximation assuming that the BEC scenario is still valid. It is showed that the anomalous density is comparable to the condensed one and survives at temperatures exceeding T_c where the condensate fraction is zero. We predicted that the shift of the critical temperature T_c due to a finite exchange anisotropy is rather substantial even when the anisotropy parameter γ is small, e.g., $\Delta T_c \approx 10$ % of T_c in H=6 T and for $\gamma \approx 4meV$. We also predicted a plausible value for the critical exponent $\beta = 0.47$ for satggered magnetization $M_\perp = \text{const} \times (1 - T/T_c)^\beta$.

2. We obtained the thermodynamic potential Ω of a triplon gas taking into account the strength of DM interaction up to second order. It was shown that the phase angle of a purely homogenous BEC without any anisotropy may only take values $\theta = \pi n$ $(n = 0, \pm 1, \pm 2 \ldots)$ while that of BEC with even a tiny DM interaction results in $\theta = \pi/2 + 2\pi n$. In contrast to the widely used Hartree-Fock-Popov approximation, which allows arbitrary phase angle, our approach predicts that the phase angle may have only discrete values, while the phase of the wave function of the whole system remains arbitrary as expected.

3. Having fixed the problem about the phases we studied the influence of possible phases to the interference and Josephson junction of two Bose condensates. We have shown that when one of the condensates have even a tiny DM interaction the interference picture will change drastically. Analyzing the simple d.c. Josephson effect between two spin-gapped magnets we have found that there would be no Josephson

current when neither of samples has DM anisotropy $(\gamma'_1 = 0, \gamma'_2 = 0)$, while the current will be finite, when one of them has a weak DM anisotropy $(\gamma'_1 = 0, \gamma'_2 \neq 0)$.

4. We have also shown the consequences of DM interaction to possible direction of staggered magnetization in spin-gapped antiferromagnets. It was shown that DM interactions destroy the total staggered magnetization in axially symmetric samples compared with $\gamma' = 0$. In the presence of DM interaction $(\gamma' \neq 0)$, the staggered magnetization remains finite. In quantum magnets, there is no path from a pure BEC to normal phase and there is a crossover instead of second order phase transition. It was shown that, in contrast to exchange anisotropy interaction, the Dzyaloshinsky-Moriya interaction modifies the physics dramatically. Particularly, it changes the sign of the anomalous density to opposite in the whole range of temperatures and changes the shape of the specific heat.

5. We also gave a fair theoretical description of the staggered magnetization data for $T \leq T_c \leq T$. By 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 studied and found to develop a linear dispersion similar to Goldstone mode with a negligibly small anisotropy gap. We come to the conclusion that to describe existing experimental data on magnetization as well as the energy spectrum of spin gapped quantum magnets with anisotrophies simultaneously, one has to extend this approach, by choosing the vector of DM anisotropy appropriately.

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Abbreviations

SSB - Spontaneous breaking of symmetry

QPT - Quantum phase transitions

MFA - Mean - field approximation

BEC - Bose-Einstein condensation

EA - Exchange anisotropy

DM - Dzyaloshinskii-Moriya

INS - Inelastic neutron scattering

ESR - Electron spin resonance

HFP - Hartree-Fock-Popov

HFB - Hartee-Fock-Bogolyubov

ESB - Explicitly breaking of symmetry

Some constants and definitions

In this work we asume the following simplifications:

$\hbar = 1$ for the Planck constant, $k_B = 1$ for the Boltzmann constant, and $V = 1$ for the unit cell volume. The unit of mass m is given in K $^{-1}$, the energy is measured in Kelvin, magnetic fields are measured in Tesla (T). The momentum and heat capacity C_H are dimensionless.

Appendix

Appendix A: Dispersion relation

The summation by momentums may be explicitly written as

$$\sum_{\mathbf{k}} f(\varepsilon(k)) = \frac{1}{(2\pi)^3}\begin{cases} 4\pi\int_0^\infty k^2 dk\varepsilon(k), \quad \varepsilon(k) = \frac{k^2}{2m} & , \\ \int_{-\pi}^{\pi} dk_x dk_y dk_z \varepsilon(k), \quad \varepsilon(k) - isotropic & , \\ \frac{1}{2}\int_{-\pi}^{\pi} dk_x \int_{-\pi}^{\pi} dk_y \int_{-2\pi}^{2\pi} dk_z \varepsilon(k), \quad \varepsilon(k) - anisotropic. \end{cases}$$

(A.1)

The isotropic bare dispersion may be presented as $\varepsilon(k) = J[3 - \cos k_x a - \cos k_y a - \cos k_z a]$ where a is the size of the unit cell (below we set $a = 1$), while the anisotropic one may be written as

$$\begin{aligned} \varepsilon_{k-k_0} &= -\Delta_{st} + \sqrt{(J+\tilde{a})^2 - \tilde{a}^2}, \\ \tilde{a} &= J_a \cos(k_x) + J_{a2c}\cos(2k_x + k_z) + 2J_{abc}\cos(k_x + k_z/2)\cos(k_y/2). \end{aligned} \quad \text{(A.2)}$$

In practical calculations with this realistic dispersion one may make a shift as $\mathbf{k} - \mathbf{k}_0 \to \mathbf{k}$, so that $\varepsilon(k-k_0)|_{k=k_0} \to \varepsilon(k)|_{k=0} = 0$, $k_0 = \{0,0,2\pi\}$ and introducing $q_x = k_x/\pi$, $q_y = k_y/\pi$, $q_z = k_z/4\pi$ we can rewrite the summation as

$$\sum_{\mathbf{k}} f(\varepsilon(k))|_{aniz} = \frac{1}{2}\int_{-1}^{1} dq_x \int_0^1 dq_y \int_0^1 dq_z f(\varepsilon(q)), \quad \text{(A.3)}$$

where $\varepsilon_q = -\Delta_{st} + \sqrt{J^2 + 2Ja_q}$, and

$$a_q = J_a\cos(\pi q_x) + J_{a2c}\cos(2\pi q_x - 4\pi q_z) - 2J_{abc}\cos(\pi q_x - 2\pi q_z)\cos(\pi q_y/2).$$

(A.4)

The condition $\varepsilon_q(q=0)=0$ fixes Δ_{st} as $\Delta_{st}=\sqrt{J^2+2J(J_a+J_{a2c}-2J_{abc})}$. In the present work we used the following values of parameters : $J=63.7$ K, $J_a=-2.5$ K, $J_{a2c}=-18.35$ K and $J_{abc}=5.28$ K, so that $\Delta_{st}=7.55$ K.

Appendix B: Calculation of the grand Hamiltonian

In this appendix we outline the derivation of the Hamiltonian (1.4) and (1.5) from the basic spin Hamiltonian and present the derivation of Ω. Taking into account the dimerized nature of the ground state, we consider a Heisenberg Hamiltonian given by

$$H = \sum_i J_0 S_{i1} \cdot S_{i2} + \sum_i \sum_{m=1}^{9} J_m S_{i,1} \cdot S_{i+\delta r_m,1} \tag{B.1}$$

where i denotes locations of dimers. J_0 indicates the intradimer spin coupling while interdimer spin couplings are described by J_m (m = 1, . . . , 9). The two spins within a dimer are labeled by subscripts 1 and 2. Since a dimer is made of two neighboring spins , the interaction between two dimers contains four different spin couplings. Now following bond-operator representation we introduce following transformations:

$$S_{1\alpha} = \frac{1}{2}\left(s^{\dagger} t_{\alpha} + t_{\alpha}^{\dagger} s - i\epsilon_{\alpha\beta\gamma} t_{\beta}^{\dagger} t_{\gamma}\right), \quad S_{2\alpha} = \frac{1}{2}\left(-s^{\dagger} t_{\alpha} - t_{\alpha}^{\dagger} s - i\epsilon_{\alpha\beta\gamma} t_{\beta}^{\dagger} t_{\gamma}\right) \tag{B.2}$$

with $\alpha = x, y, z$ and $\epsilon_{\alpha\beta\gamma}$ is the totally antisymmetric unit tensor. The corresponding auxiliary quasiparticles can, therefore, be called singletons, and triplons, respectively. The restriction on the physical states to be either singlets or triplets leads to the constraint

$$s^{\dagger} s + \sum_{\alpha} t_{\alpha}^{\dagger} t_{\alpha} = 1 \tag{B.3}$$

Inserting (B.2) into the spin Hamiltonian (B.1) and using constraints (B.3) one may obtain effective Bose Hamiltonian (1.4) and (1.5).

In the notion of representative ensemble the grand Hamiltonian including the exchange anisotropy term can be written as:

$$H = \hat{H} - \mu_0 \hat{N}_0 - \mu_1 \hat{N}_1 - \hat{\Lambda},$$
$$\hat{H} = \int \left\{ \psi^\dagger(\mathbf{r}) \hat{K} \psi(\mathbf{r}) + \frac{U}{2} \left(\psi^\dagger(\mathbf{r}) \psi(\mathbf{r}) \right)^2 + \frac{\gamma}{2} \left(\psi^\dagger(\mathbf{r}) \psi^\dagger(\mathbf{r}) + \psi(\mathbf{r}) \psi(\mathbf{r}) \right) \right\} d^3 r, \tag{B.4}$$

where $\hat{N}_0 = \int |\phi_0|^2 d^3 r$, $\hat{N}_1 = \int \tilde{\psi}^\dagger(\mathbf{r}) \tilde{\psi}(\mathbf{r}) d^3 r$, so that $\mu N = \mu_0 N_0 - \mu_1 N_1$, $\hat{N} = \int \psi^\dagger \psi d^3 r$ is the total number of particles. The Lagrange multiplier

$$\hat{\Lambda} = \int [\lambda \tilde{\psi}^\dagger(\mathbf{r}) + \lambda^\dagger \tilde{\psi}(\mathbf{r})] d^3 r \tag{B.5}$$

is a so called linear killer, such that λ is chosen from the constraint for the conservation of quantum numbers, $\langle \tilde{\psi}(\mathbf{r}) \rangle = 0$. The quantum fluctuation $\tilde{\psi}(\mathbf{r})$ is related to the field operator as $\psi(\mathbf{r}) = \phi_0 + \tilde{\psi}(\mathbf{r})$, which makes possible to rewrite the grand Hamiltonian as follows:

$$H = H_0 + H_1 + H_2 + H_3 + H_4, \tag{B.6}$$

where

$$H_0 = \int d^3 r \rho_0 (-\mu_0 + \gamma + \frac{U}{2} \rho_0) \tag{B.7}$$

with $\rho_0 = \phi_0^2$

$$H_1 = \int d^3 r \phi_0 (\gamma + U\rho_0)(\tilde{\psi}^\dagger(\mathbf{r}) + \tilde{\psi}(\mathbf{r})), \tag{B.8}$$

$$H_2 = \int d^3 r \left\{ \tilde{\psi}^\dagger(\mathbf{r}) [\hat{K} - \mu_1 + 2U\rho_0] \tilde{\psi}(\mathbf{r}) + \frac{1}{2} \left(\tilde{\psi}^2(\mathbf{r}) + \tilde{\psi}^{\dagger 2}(\mathbf{r}) \right) (\gamma + U\rho_0) \right\}, \tag{B.9}$$

$$H_3 = U\phi_0 \int d^3 r \tilde{\psi}^\dagger(\mathbf{r}) \tilde{\psi}(\mathbf{r}) \left(\tilde{\psi}^\dagger(\mathbf{r}) + \tilde{\psi}(\mathbf{r}) \right), \tag{B.10}$$

$$H_4 = \frac{U}{2} \int d^3 r (\tilde{\psi}^\dagger(\mathbf{r}) \tilde{\psi}(\mathbf{r}))^2. \tag{B.11}$$

Performing a Fourier transformation $\tilde{\psi}(\mathbf{r}) = \sum_{\mathbf{k}\neq 0} a_{\mathbf{k}}\tilde{\psi}_{\mathbf{k}}(\mathbf{r})$ and assuming that $\phi_0(\mathbf{r})$ does not depend on $\mathbf{r}$ we may rewrite the above equations as follows:

$$H_0 = \rho_0(-\mu_0 + \gamma + \frac{U}{2}\rho_0), \tag{B.12}$$

$$H_1 = \sqrt{\rho_0}(U\rho_0 + \gamma)\sum_{\mathbf{k}} (a_{\mathbf{k}} + a_{\mathbf{k}}^{\dagger}), \tag{B.13}$$

$$H_2 = \sum_{\mathbf{k}} \{\varepsilon_k - \mu_1 + 2U\rho_0\}a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}} + \frac{1}{2}(a_{\mathbf{k}}a_{-\mathbf{k}} + a_{\mathbf{k}}^{\dagger}a_{-\mathbf{k}}^{\dagger})(U\rho_0 + \gamma), \tag{B.14}$$

$$H_3 = U\sqrt{\rho_0}\sum_{\mathbf{k},\mathbf{p}} (a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}+\mathbf{p}}a_{-\mathbf{p}} + \mathrm{h.c.}), \tag{B.15}$$

$$H_4 = \frac{U}{2}\sum_{\mathbf{k},\mathbf{q},\mathbf{p}} a_{\mathbf{k}}^{\dagger}a_{\mathbf{p}}^{\dagger}a_{\mathbf{p}+\mathbf{q}}a_{\mathbf{k}-\mathbf{q}}. \tag{B.16}$$

The third and fourth order terms may be further simplified by using the approximation given in Eq. (1.17) as:

$$\begin{aligned} H_3 &= U\sqrt{\rho_0}(2\rho_1 + \sigma)\sum_{\mathbf{k}} (a_{\mathbf{k}}^{\dagger} + a_{\mathbf{k}}), \\ H_4 &= \frac{U}{2}\{4\rho_1 a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}} + \sigma(a_{\mathbf{k}}a_{-\mathbf{k}} + a_{-\mathbf{k}}^{\dagger}a_{\mathbf{k}}^{\dagger}) - (2\rho_1^2 + \sigma^2)\}. \end{aligned} \tag{B.17}$$

Now the grand Hamiltonian is the sum of classical, H_{class}, linear H_{lin} and H_{bilin} terms as:

$$\begin{aligned} H &= H_{\mathrm{class}} + H_{\mathrm{lin}} + H_{\mathrm{bilin}}, \\ H_{\mathrm{class}} &= -\rho_0\mu_0 + \rho_0\gamma + \frac{U}{2}\rho_0^2 - \frac{U}{2}(2\rho_1^2 + \sigma^2), \\ H_{\mathrm{lin}} &= U\sqrt{\rho_0}(\rho_0 + \tilde{\gamma} + 2\rho_1 + \sigma)\sum_{\mathbf{k}} (a_{\mathbf{k}}^{\dagger} + a_{\mathbf{k}}), \\ H_{\mathrm{bilin}} &= \sum_{\mathbf{k}} (\varepsilon_k - \mu_1 + 2U\rho)a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}} + \frac{U(\rho_0 + \tilde{\gamma} + \sigma)}{2}\sum_{\mathbf{k}} (a_{\mathbf{k}}a_{-\mathbf{k}} + a_{\mathbf{k}}^{\dagger}a_{-\mathbf{k}}^{\dagger}), \end{aligned} \tag{B.18}$$

where $\tilde{\gamma} = \gamma/U$ and $\rho = \rho_0 + \rho_1$. In the formalism of representative ensemble the linear term is neglected by an appropriate choice of λ, for example, by choosing $\lambda = U\sqrt{\rho_0}(\rho_0 + \tilde{\gamma} + 2\rho_1 + \sigma)$ in Eq. (B.5) . The μ_0 can be found by minimization of the free energy with respect to ρ_0. To diagonalize the bilinear term we introduce the normal Σ_n and anomalous Σ_{an} self energies as

$$\Sigma_n = 2U\rho, \qquad \Sigma_{an} = U(\rho_0 + \tilde{\gamma} + \sigma), \tag{B.19}$$

such that H_{bilin} is rewritten as

$$H_{\text{bilin}} = \sum_{\mathbf{k}} \omega_k a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \frac{\Sigma_{an}}{2} \sum_{\mathbf{k}} (a_{\mathbf{k}} a_{-\mathbf{k}} + a_{\mathbf{k}}^\dagger a_{-\mathbf{k}}^\dagger), \tag{B.20}$$

where

$$\omega_k = \varepsilon_k - \mu_1 + \Sigma_n. \tag{B.21}$$

The next step is the Bogolyubov transformation

$$a_{\mathbf{k}} = u_{\mathbf{k}} b_{\mathbf{k}} + v_{\mathbf{k}} b_{-\mathbf{k}}^\dagger, \quad a_{\mathbf{k}}^\dagger = u_{\mathbf{k}} b_{\mathbf{k}}^\dagger + v_{\mathbf{k}} b_{-\mathbf{k}} \tag{B.22}$$

to diagonalize H_{bilin}. The operators $b_{\mathbf{k}}$ and $b_{\mathbf{k}}^\dagger$ can be interpreted as annihilation and creation operators of phonons with following properties:

$$[b_{\mathbf{k}}, b_{\mathbf{p}}^\dagger] = \delta_{\mathbf{k},\mathbf{p}}, \quad \langle b_{\mathbf{k}}^\dagger b_{-\mathbf{k}}^\dagger \rangle = \langle b_{\mathbf{k}} b_{-\mathbf{k}} \rangle = 0, \tag{B.23}$$

$$\langle b_{\mathbf{k}}^\dagger b_{\mathbf{k}} \rangle = f_B(E_k) = \frac{1}{e^{\beta E_k - 1}}, \tag{B.24}$$

where $\beta \equiv 1/T$. To determine the phonon dispersion E_k we insert (B.22) into (B.20) and require that the coefficient of the term $b_{\mathbf{k}} b_{-\mathbf{k}} + b_{-\mathbf{k}}^{\dagger} b_{\mathbf{k}}^{\dagger}$ vanishes, i.e:

$$\omega_k u_{\mathbf{k}} v_{\mathbf{k}} + \frac{\Sigma_{an}}{2}\left(u_{\mathbf{k}}^2 + v_{\mathbf{k}}^2\right) = 0. \tag{B.25}$$

Now using the condition $u_{\mathbf{k}}^2 - v_{\mathbf{k}}^2 = 1$ and presenting $u_{\mathbf{k}}, v_{\mathbf{k}}$ as

$$u_{\mathbf{k}}^2 = \frac{\omega_k + E_k}{2E_k}, \qquad v_{\mathbf{k}}^2 = \frac{\omega_k - E_k}{2E_k} \tag{B.26}$$

yields

$$\sqrt{\omega_k^2 - E_k^2} = -\Sigma_{an}, \qquad u_{\mathbf{k}} v_{\mathbf{k}} = -\frac{\Sigma_{an}}{2E_k}, \qquad u_{\mathbf{k}}^2 + v_{\mathbf{k}}^2 = \frac{\omega_k}{E_k} \tag{B.27}$$

that is

$$E_k^2 = (\omega_k + \Sigma_{an})(\omega_k - \Sigma_{an}) \equiv (\varepsilon_k + X_1)(\varepsilon_k + X_2) \tag{B.28}$$

with

$$X_1 = \Sigma_n + \Sigma_{an} - \mu_1, \qquad X_2 = \Sigma_n - \Sigma_{an} - \mu_1 \tag{B.29}$$

Now H_{bilin} is simplified as

$$H_{\text{bilin}} = \sum_{\mathbf{k}} E_k b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} + \frac{1}{2}\sum_{\mathbf{k}} (E_k - \omega_k) \tag{B.30}$$

and the total Hamiltonian is given by

$$H = H_{\text{class}} + H_{\text{bilin}} = -\rho_0\mu_0 + \rho_0\gamma + \frac{U}{2}\rho_0^2 - \frac{U}{2}(2\rho_1^2 + \sigma^2) + \sum_{\mathbf{k}} E_k b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} + \frac{1}{2}\sum_{\mathbf{k}} (E_k - \omega_k) \tag{B.31}$$

which may be used to define the energy of the system.