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Magnon Bose condensation in a symmetry breaking magnetic field

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Abstract

Magnon Bose condensation (BC) in a symmetry breaking magnetic field is the result of an unusual form of Zeeman energy that has terms linear in the spin-wave operators and terms mixing excitations, the momenta of which differ in the wavevector of the magnetic structure. The following examples are considered: simple easy-plane tetragonal antiferromagnets (AFs), the frustrated AF family R_2CuO_4, where R $=$ Pr, Nd etc, and cubic magnets with a Dzyaloshinskii–Moriya interaction (MnSi etc). In all cases BC is important when the magnetic field is comparable with the spin-wave gap. The theory is illustrated by existing experimental results.

1. Introduction

Magnon Bose condensation (BC) in a magnetic field has been intensively studied in spin singlet materials (see, for example, [1] and references therein). In this case magnons condense in the field just above the triplet gap. In this paper we consider magnon BC that appears in a symmetry breaking magnetic field. The theoretical discussion is illustrated by experimental observation of this BC in frustrated antiferromagnet (AF) Pr_2CuO_4 and the cubic helimagnets MnSi and FeGe. To clarify our idea we begin by considering conventional AFs. In textbooks two limiting cases are considered. The first case is when the magnetic field is directed along the sublattices. In this case the system remains stable up to the critical field $H_C = \Delta$, where Δ is the spin-wave gap. Then a first order transition occurs to the state in which the field is perpendicular to the sublattices (spin–flop transition). In the second case the field is perpendicular to the initial staggered magnetization. The system remains stable but the spins are canted toward the field by the angle determined by $\sin\vartheta = -H/(2SJ_0)$, where $J_0 = Jz$ and J and z are the exchange interaction and the number of nearest neighbours, respectively. At $H + 2SJ_0$ a spin–flip transition occurs to the ferromagnetic state. To the best of our knowledge the first consideration of the symmetry breaking field was made theoretically in [2] in connection with an experimental study of the magnetic structure of the frustrated AF R_2CuO_4, where R $=$ Pr,

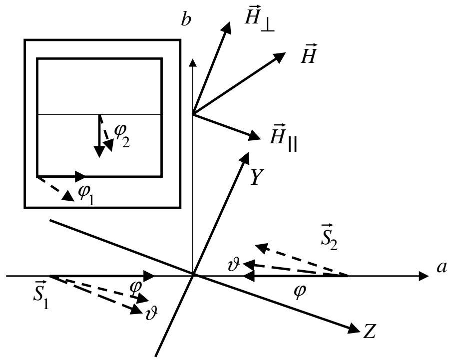


Figure 1. Spin configuration in the field. Full and dashed arrows correspond to zero and nonzero field, respectively. Additional spin canting in $H_\perp$ is shown by broken arrows. Inset: spin configuration in neighbouring planes of a frustrated AF.

Nd, Sm and Eu [3, 4]. In these papers a non-collinear structure was observed using neutron scattering in the field directed at an angle of $\delta = 45°$ to the sublattices. It was found in [2] that in an inclined field the Zeeman energy has an unusual form with terms which are linear in the spin-wave operators and terms mixing magnons with momenta which differ in the AF vector $\boldsymbol{k}_0$. As a result there arises BC of the spin-waves with momenta equal to zero and $\pm\boldsymbol{k}_0$.

A similar situation exists in cubic helimagnets, MnSi etc [5]. If the field is directed along the helical wavevector $\boldsymbol{k}$ the plain helix transforms into a conical structure and then the ferromagnetic spin state occurs at a critical field H_C. But if $\boldsymbol{H} \perp \boldsymbol{k}$ the magnons condense with momenta zero, $\pm\boldsymbol{k}$, $\pm 2\boldsymbol{k}$ etc. This leads to the following observable phenomena:

(i) a transition to the state with $\boldsymbol{k}$ directed along the field at $H_\perp \sim H_{C1} = \Delta\sqrt{2}$, where Δ is the spin-wave gap,

(ii) the second harmonic of the spin rotation with the vector $2\boldsymbol{k}$ and perpendicular spin susceptibility at $H_\perp < H_{C1}$. Rotation of the helix was observed in [6–8].

In this paper we outline basic ideas about BC in a symmetry breaking field as applied to frustrated cuprates and non-centrosymmetric cubic helimagnets. We illustrate the theory with some recent experiments.

2. Non-frustrated AF

To demonstrate the basic ideas of our approach we begin with non-frustrated easy-plane tetragonal AF. We are not interested in thermal fluctuations and consider a single AF plane. If the field is directed at an angle δ to the b axis (see figure 1) sublattices rotate by an angle φ. Simultaneously, the field component perpendicular to the new Z axis cants the spins by an angle $\vartheta \simeq -H_\perp/(2SJ_0) \ll 1$. As a result we have for the two neighbouring spins in (ZY) frame [2]

$$\boldsymbol{S}_1 = S_{z1}\hat{Z} + \vartheta S_{y1}\hat{Y}; \qquad \boldsymbol{S}_2 = -S_{z2}\hat{Z} + \vartheta S_{y2}\hat{Y}, \tag{1}$$

where in the linear spin-wave theory $S_{zl} = S - a_l^+ a_l$ and $S_{yl} = -\mathrm{i}\sqrt{S/2}(a_l - a_l^+)$, $l = 1, 2$ and $a_l(a_l^+)$ are Bose operators. As a result the Zeeman energy has the unusual form

$$H_Z = H_\| \sum a^+_{\boldsymbol{q}+\boldsymbol{k}_0} a_{\boldsymbol{q}} + \mathrm{i}\vartheta\sqrt{NS/2}(a_0 - a_0^+), \tag{2}$$

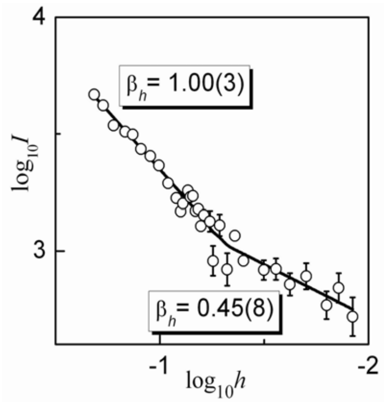


Figure 2. Log–log plot of the $(1/2, 1/2, -1)$ Bragg intensity in a diagonal field, $h \sim (H_C - H)$.

where $\boldsymbol{k}_0$ and N are the AF wavevector and total spin number, respectively. Here the first term mixes spin-waves with momenta $\boldsymbol{q}$ and $\boldsymbol{q} \pm \boldsymbol{k}_0$ and the second one excites (absorbs) magnons with $\boldsymbol{q} = 0$. Along with this energy we have conventional spin-wave Hamiltonian $H_{SW} = \sum [E_q a_q^+ a_q + B_q(a_{-q}a_q + a_{-q}^+ a_q^+)/2]$ with the spin-wave energy $\epsilon_q = (E_q^2 - B_q^2)^{1/2}$. The spin-wave gap is $\epsilon_0 = \Delta$.

Linear terms in equation (2) contribute to the ground state energy if $a_0(a_0^+) \sim \sqrt{N}$, i.e. these operators have to be considered as classical variables as in the Bogoliubov theory of the BC in a dilute Bose gas. Due to the first term in (2) we must consider the operators $a_{\pm \boldsymbol{k}_0}$ and $a_{\mp \boldsymbol{k}_0}^+$ as classical variables too. Minimizing the full Hamiltonian with respect to these variables we obtain

$$E = (\Delta^2 \sin^2 2\varphi)/(16J_0) - S^2 J_0 \vartheta^2 - (H_\parallel H_\perp)^2/[4J_0(\Delta^2(\varphi, \boldsymbol{H})], \tag{3}$$

where the first term is the energy of the square anisotropy. In cuprates with $S = 1/2$ it has a quantum origin and arises due to pseudodipolar in-plane interaction [9]. The second term is the energy of the spin canting in a perpendicular field. The last term is the BC energy and $\Delta^2(\varphi, \boldsymbol{H}) = \Delta^2 \cos 4\varphi + H_\perp^2 - H_\parallel^2$ is the spin-wave gap in the field [2]. This contribution becomes important at $H \sim \Delta$. The spin configuration is determined by $\mathrm{d}E/\mathrm{d}\varphi = 0$ and the equilibrium condition $\mathrm{d}^2E/\mathrm{d}\varphi^2 \geqslant 0$.

This theory was verified by neutron scattering [10, 11]. In a diagonal field $H \parallel (1, 1, 0)$ the spin configuration in frustrated Pr_2CuO_4 is governed by equation (3) and the intensity of the $(1/2, 1/2, -1)$ is given by $I \sim 1 + \sin 2\varphi$ [2]. Neglecting the BC term we get $\sin 2\varphi = -(H/H_C)^2$, where $H_C = \Delta$. As a result at $H \to H_C$ we obtain $I \sim H_C - H$. But very close to H_C the BC term becomes important and we have a crossover to $I \sim (H_C - H)^{1/2}$. This is clearly seen in figure 2. This crossover was observed in [10, 11].

3. Frustrated AFs

In frustrated R_2CuO_4 AFs there are two copper spins in a unit cell belonging to different CuO_2 planes (see inset in figure 1). From symmetry considerations these spins do not interact in the exchange approximation. The orthogonal spin structure is a result of the interplane pseudodipolar interaction (PDI) [2, 3] and the ground state energy is given by

$$E = \frac{\Delta^2}{16J_0}[\sin^2 2\varphi_1 + \sin^2 2\varphi_2 - 4G \sin(\varphi_1 + \varphi_2)] - S^2 J_0(\vartheta_1^2 + \vartheta_2^2) + E_C(\varphi_1, \varphi_2, \boldsymbol{H}), \tag{4}$$

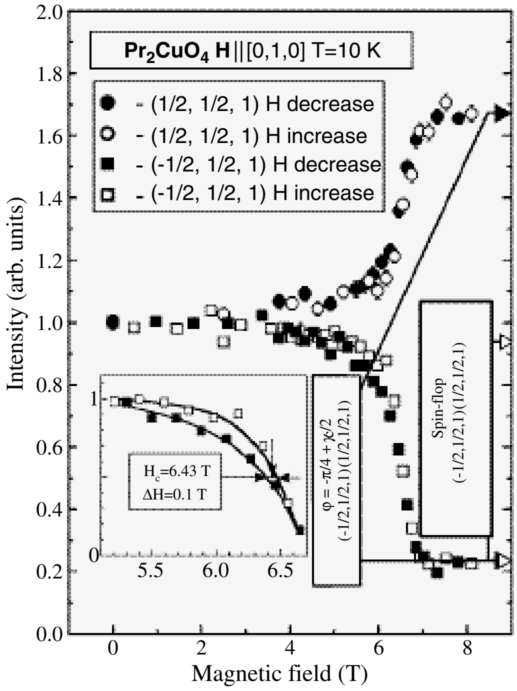


Figure 3. The first order transition in the field directed along the b axis. Calculated intensities for the spin–flop configurations when spins are perpendicular to the field (white arrows).

where $\vartheta_{12} = -H_{\perp 12}/(2SJ_0)$, $G = (\Omega/\Delta)^2$ and Ω^2 is the difference between the square of the optical and acoustic spin-wave branches at $H = 0$. The BC energy E_C has a very complicated form [2] and we do not present it here.

For Pr_2CuO_4 at $T = 18$ K we have $\Delta \simeq 0.36$ meV, $\Omega \simeq 2.8$ meV and $\Omega \simeq 60 \ll 1$ [2]. Then the intraplane PDI is strong and the BC contribution cannot be neglected at low fields $H < \Delta$. We illustrate this with the results of particular calculations including and neglecting BC in the field almost along the b axis ($\delta \ll 1$). Instead of $\varphi_{1,2}$ we use new angles determined as $\varphi_1 = \alpha + \gamma/2, \quad \varphi_2 = -\pi/2 - \alpha + \gamma/2$. Neglecting BC we have $\alpha = -(H/\Delta)^2\delta$ and $\gamma = (H/\Delta)^4\delta/G$. BC changes the last result: $\gamma_{BC} = \gamma G \gg \gamma$.

The role of BC can be illustrated by the results of neutron scattering in Pr_2CuO_4 [12]. The angles α and γ were determined from measurements of two reflections $(1/2, 1/2, 1)$ and $(-12, 1/2, 1)$. If $\delta = 0$ the zero field spin configuration remains stable at $H < H_C \Delta G^{1/4}$ and there is no BC as $H_{\parallel 1} = H_{\perp 2} = 0$ (see equation (2)). Then the theory without BC predicts a first order transition to the collinear non-spin–flop state with $\alpha = 45°$ and $\tan\gamma_C = G^{1/2}$. This transition is seen in figure 3 at $H_C \simeq 6.5$ T [12]. From these data we obtain $\gamma_C \simeq 30°$. Using parameters given above and neglecting the BC we obtain $H_C \simeq 6.7$ T and $\gamma_C \simeq 7.4°$. The last quantity is in strong disagreement with experiment. It was demonstrated in [13] that Δ depends on temperature, and at $T = 10$ K we have $\Delta \simeq 0.5$ meV as in figure 3. Assuming that Ω does not depend on T we obtain $H_C \simeq 7.8$ T and $\gamma_C \simeq 5.3°$ that is in stronger disagreement with the experiment.

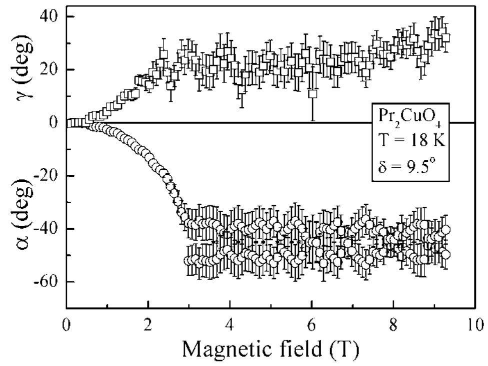


Figure 4. Field dependence of angles α and γ at $\delta = 9.5°$.

The experimentally obtained angles α and γ at $T = 18$ K and $\delta = 9.5°$ are shown in figure 4 [12]. The transition to the collinear state with $\alpha \sim -45°$ and $\gamma_C \sim 20°$ was observed. Again the non-BC theory cannot explain the experimental data. For example it gives $\gamma_C \simeq 2.5°$. An explanation of all these experimental data using the BC theory will be given elsewhere.

4. BC in helimagnets

In the helimagnets MnSi etc the Dzyaloshinskii–Moriya interaction (DMI) stabilizes the helical structure and the wavevector of the helix has the form $\boldsymbol{k} = SD[\hat{a} \times \hat{b}]/A$, where D is the strength of the DMI, A is the spin-wave stiffness at momenta $q \gg k$ and $\hat{a}$ and $\hat{b}$ are unit orthogonal vectors in the plane of the spin rotation.

The classical energy depends on the field component $H_\parallel$ along the vector $\boldsymbol{k}$ and the cone angle of the spin rotation is given by $\sin\alpha = -H/H_C$, where $H_C = Ak^2$ is the critical field of the transition into the ferromagnetic state [5]. However, at $H_\perp \ll H_C$ rotation of the axis of the helix toward the field direction and the second harmonic $2\boldsymbol{k}$ of the spin rotation was observed [6–8]. Both phenomena are related to the magnon BC in a perpendicular field [5].

The linear and mixing terms appear in the Zeeman energy in much the same way as discussed above:

$$H_Z = (H_a - \mathrm{i}H_b)\sqrt{NS/2}(a_{-\boldsymbol{k}} - a^+_{\boldsymbol{k}})/2 - \sum(a^+_{\boldsymbol{k}} a_0 + a^+_0 a_{-\boldsymbol{k}}) + \mathrm{H.C.}, \tag{5}$$

and we have magnon BC with momenta zero and $\pm\boldsymbol{k}$. The corresponding contribution to the ground state energy is given by

$$E_C = -SH^2_\perp \Delta^2/[H_C(\Delta^2 - H^2_\perp/2)]. \tag{6}$$

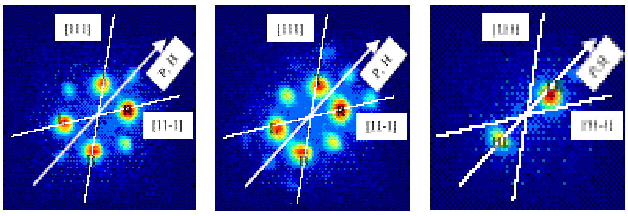
Obviously near the critical point $H_\perp = \Delta\sqrt{2}$ the real form of the BC energy is not so simple. It is determined by nonlinear interactions, but consideration of this problem is outside the scope of this paper.

As a result the perpendicular susceptibility is proportional to $1/(\Delta^2 - H^2_\perp/2)$ and a $2\boldsymbol{k}$ harmonic appears. The latter was observed by neutron scattering [6–8]. The intensities of the corresponding Bragg satellites have the form

$$I_\pm \sim [\Delta^2/(\Delta^2 - H^2_\perp/2)]^2[1 \mp (\boldsymbol{kP})]\delta(\boldsymbol{q} \mp 2\boldsymbol{k}), \tag{7}$$

where $\boldsymbol{P}$ is the neutron polarization.

If $H_\perp \to \Delta\sqrt{2}$ the axis of the helix rotates toward the field. This rotation is governed by competition of the BC and crystallographic energies [5]. Evolution of the Bragg reflections in MnSi with $H_\perp$ is shown in figure 5.



a)$H = 10mT$ *b*)$H = 50mT$ *c*)$H = 150mT$

Figure 5. Bragg reflections in the field along $(1, 1, 0)$. (a) Four strong spots corresponds to $\pm(1, 1, 1)$ and $\pm(1, 1, -1)$ reflections. Weak spots are the double Bragg scattering. (b) The $2\boldsymbol{k}$ satellites appear. (c) The helix vector is directed along the field.

(This figure is in colour only in the electronic version)

5. Conclusions

We discussed a few examples of magnon BC in a symmetry breaking magnetic field. BC appears to be due to unusual terms in the Zeeman energy. Obviously this phenomenon is very general and can be observed in other ordered magnetic systems. Effects related to BC are more pronounced in a field of the order of the sine-wave gap.

Acknowledgments

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