Phase Diagram for Magnon Condensate in Yttrium Iron Garnet film

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Recently, magnons, which are quasiparticles describing the collective motion of spins, were found to undergo Bose-Einstein condensation (BEC) at room temperature in films of Yttrium Iron Garnet (YIG). Unlike other quasiparticle BEC systems, this system has a spectrum with two degenerate minima, which makes it possible for the system to have two condensates in momentum space. Recent Brillouin Light scattering studies for a microwave-pumped YIG film of thickness $d = 5$ μm and field $H = 1$ kOe find a low-contrast interference pattern at the characteristic wavevector Q of the magnon energy minimum. In this report, we show that this modulation pattern can be quantitatively explained as due to non-symmetric but coherent Bose-Einstein condensation of magnons into the two energy minima. Our theory predicts a transition from a high-contrast symmetric phase to a low-contrast non-symmetric phase on varying the d and H, and a new type of collective oscillations.

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Bose-Einstein condensation (BEC), one of the most intriguing macroscopic quantum phenomena, has been observed in equilibrium systems of Bose atoms, like ^{4}He [1, 2], ^{87}Rb [3] and ^{23}Na [4]. Recent experiments have extended the concept of BEC to non-equilibrium systems consisting of photons [5] and of quasiparticles, such as excitons [6], polaritons [7–9] and magnons [10, 11]. Among these, BEC of magnons in films of Yttrium Iron Garnet (YIG), discovered by the group of Demokritov [11–17], is distinguished from other quasiparticle BEC systems by its room temperature transition and two-dimensional anisotropic properties. In particular, the spin-wave energy spectrum of a YIG film shows two energetically degenerate minima. Therefore it is possible that the system may have two condensates in momentum space [18]. An experiment by Nowik-Boltyk *et al.* [17] indeed shows a low-contrast spatial modulation pattern, indicating that there is interference between the two condensates. Current theories [19–24] do not describe the appearance of coherence or the distribution of the two condensates.

This report points out that a complete description of BEC in microwave-pumped YIG films must account for the 4th order interactions, including previously neglected magnon-non-conserving terms originating in the dipolar interactions. The theory explains not only the appearance of coherence but also quantitatively explains the low contrast of the experimentally observed interference pattern. Moreover, the theory predicts that, on increasing the film thickness d from a small value, there is a transition from a high-contrast symmetric phase S for $d < d_c$, with equal numbers of condensed magnons filling the two minimum states, to a low-contrast coherent non-symmetric phase NS for $d > d_c$, with different numbers of condensed magnons filling the two minimum states. In comparatively thin films ($d < 0.2\mu$m) the same transition can be driven by an external magnetic field H. For $d > d^*$, where d^* is another critical thickness ($d^* > d_c$), the sum of phases of the two condensates changes from π to 0; for $d = d^*$ the system is in a completely non-symmetric phase with only one condensate, for which there is no interference. In the experiment of Ref.[17] the film thickness d exceeded d^*. We suggest that the phase transitions may be identified by measuring the contrast of the spatial interference pattern for various d and H. We also predict a new type of collective magnetic oscillation in this system and discuss the possibility of domain walls in non-symmetric phases.

Results

Phase Diagram.— We consider a YIG film of thickness d with in-plane magnetic field H (see inset of Fig. 1). The 4-th order interaction of con-

densate amplitudes reads [25–27]:

$$\hat{V}_4 = A[c_Q^\dagger c_Q^\dagger c_Q c_Q + c_{-Q}^\dagger c_{-Q}^\dagger c_{-Q} c_{-Q}]$$
$$+ 2B c_Q^\dagger c_{-Q}^\dagger c_{-Q} c_Q$$
$$+ C[c_Q^\dagger c_Q c_Q c_{-Q} + c_{-Q}^\dagger c_{-Q} c_{-Q} c_Q + h.c.]. \quad (1)$$

Here $c_{\pm Q}$ and $c_{\pm Q}^\dagger$ are the annihilation and creation operators for magnons in the two condensates located at the two energy minima $(0, \pm Q)$ in the 2-D momentum space (see. Fig.1). The coeffi-
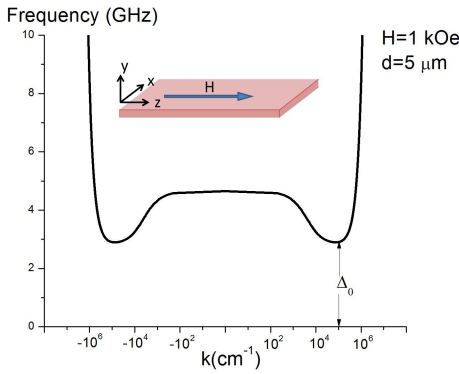

FIG. 1. The magnon spectrum in the k_z direction for $d = 5$ μm and $H = 1$ kOe. The inset is a schematic diagram of YIG film.

cients in Eq.(1) are:

$$A = -\frac{\hbar \omega_M}{4SN}[(\alpha_1 - \alpha_3)F_Q - 2\alpha_2(1 - F_{2Q})]$$
$$- \frac{DQ^2}{2SN}[\alpha_1 - 4\alpha_2],$$
$$B = \frac{\hbar \omega_M}{2SN}[(\alpha_1 - \alpha_2)(1 - F_{2Q}) - (\alpha_1 - \alpha_3)F_Q]$$
$$+ \frac{DQ^2}{SN}[\alpha_1 - 2\alpha_2],$$
$$C = \frac{\hbar \omega_M}{8SN}[(3\alpha_1 - \frac{20}{3}\alpha_3 + 3\alpha_2)F_Q$$
$$+ \frac{16}{3}\alpha_3(1 - F_{2Q})] + \frac{DQ^2}{SN}\alpha_3, \quad (2)$$

with $\alpha_1 = u^4 + 4u^2v^2 + v^4$, $\alpha_2 = 2u^2v^2$ and $\alpha_3 = 3uv(u^2 + v^2)$. Here, u and v are the coefficients of Bogoliubov transformation (see the Methods section for details). $S = 14.3$ is the effective spin, N is the total number of spins in the film, M is the magnetization and $\hbar \omega_M = \gamma 4\pi M$ with gyromagnetic

ratio $\gamma = 1.2 \times 10^{-5}$eV/kOe. D is proportional to the exchange constant and $F_k = (1 - e^{-kd})/kd$. Similar results for the coefficients A and B were obtained in Ref.[19]. The coefficient C, which violates magnon number conservation, has not been considered previously. Below we show that C is the only source of coherence between the two condensates. The three coefficients A, B and C, whose values as functions of H are shown in Fig.2 for two typical values of d, determine the distribution of condensed magnons in the two degenerate minima. Ref.[19] assumed a symmetric phase with condensed magnons in both minima having equal amplitudes and equal phases. Later, Ref.[20] assumed filling of only one minimum. More recently Ref.[24] considered Josephson-like oscillations by starting from two condensates with equal numbers of magnons but different phases. Our theory predicts coherent condensates and the ratio of their amplitudes with no additional assumptions.

In terms of condensate numbers $N_{\pm Q}$ and phases $\phi_\pm$, the condensate amplitudes are $c_{\pm Q} = \sqrt{N_{\pm Q}}e^{i\phi_\pm}$. Substituting them into eq.(1) we find:

$$V_4 = A(N_Q^2 + N_{-Q}^2) + 2BN_Q N_{-Q}$$
$$+ 2C\cos\Phi(N_Q^{\frac{3}{2}}N_{-Q}^{\frac{1}{2}} + N_Q^{\frac{1}{2}}N_{-Q}^{\frac{3}{2}}). \quad (3)$$

Clearly the dipole energy depends on the total phase $\Phi = \phi_+ + \phi_-$. To minimize this energy, for $C > 0$ we have $\Phi = \pi$ and for $C < 0$ we have $\Phi = 0$. Fig.2 shows that the sign of C changes for different d and H, which indicates a transition of Φ between 0 and π. For both $C > 0$ and $C < 0$ the dipole energy establishes a coherence between the two condensate amplitudes. In contrast to a Josephson-like interaction, the sum rather than the difference of the two condensate phases is fixed.

Since the total number of condensed magnons $N_c = N_Q + N_{-Q}$ is uniquely determined by the pumping (see Methods), the energy has only a single free variable, the so far unspecified difference $\delta = N_Q - N_{-Q}$. In terms of N_c and δ the condensate energy eq.(3) is:

$$V_4 = \frac{1}{2}\Big[(A + B)N_c^2 - (B - A)\delta^2$$
$$- 2|C|N_c\sqrt{N_c^2 - \delta^2}\Big]. \quad (4)$$

The ground state of the condensates depends on the criterion parameter Δ, defined as
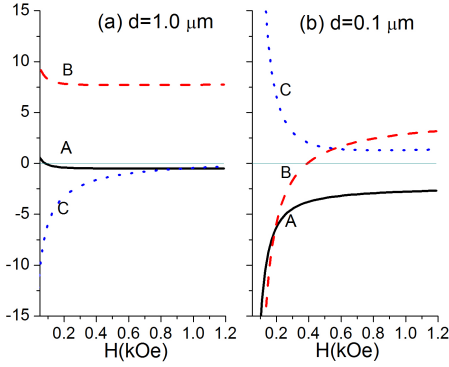
$$\Delta \equiv A - B + |C|. \quad (5)$$

FIG. 2. Interaction coefficients A, B and C (in units of mK/N, with N the total number of spins in the film) as a function of magnetic field H for film thickness (a) $d = 1.0$ μm and (b) $d = 0.1$ μm.

FIG. 3. Transition criterion Δ from non-symmetric to symmetric phase, Δ (in units of mK/N), as a function of magnetic field H for different thicknesses d.

When $\Delta > 0$, $\delta = 0$ minimizes the energy, so the two minima have equal numbers of condensed magnons. This is the symmetric phase, with $N_Q = N_{-Q}$. When $\Delta < 0$, the minimum shifts to $\frac{\delta^2}{N_c^2} = 1 - \frac{C^2}{(B-A)^2}$. This is the non-symmetric phase (S), with $N_Q \neq N_{-Q}$. The transition from symmetric to non-symmetric phase (NS) at $\Delta = 0$ is of the second order. There is no metastable state of these phases. At $C = 0$ one finds $\delta = \pm N_c$, which corresponds to a completely non-symmetric phase with only one condensate. The ground state of the non-symmetric phase is doubly-degenerate, corresponding to the two possible signs for δ. Fig.3 shows that for a film thickness of about 0.05 μm, the symmetric phase is energy favorable up to $H = 1.2$ T. For $d = 0.08$ μm, on increasing H to about 0.6 kOe, there is a transition from symmetric to non-symmetric phase. For the larger thicknesses $d = 0.1$ μm and $d = 1$ μm, the ground state is non-symmetric for $H > 0.3$ kOe.

Fig.4 gives the phase diagram in (d, H) space. It has three different regions, separated by two critical transition lines, $d_c(H)$ and $d^*(H)$, corresponding to $\Delta = 0$ and $C = 0$, respectively. For $d = 0.13 - 0.16$ μ the system possesses re-entrant behavior (NS $\Phi = \pi$, to NS $\Phi = 0$, to NS $\Phi = \pi$) as H increases. As shown below, measurement of the contrast, or modulation depth [17], of the spatial interference pattern permits identification of the different condensate phases.
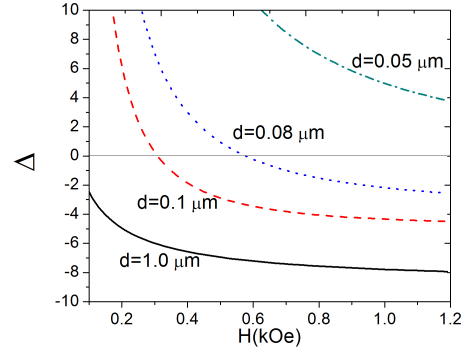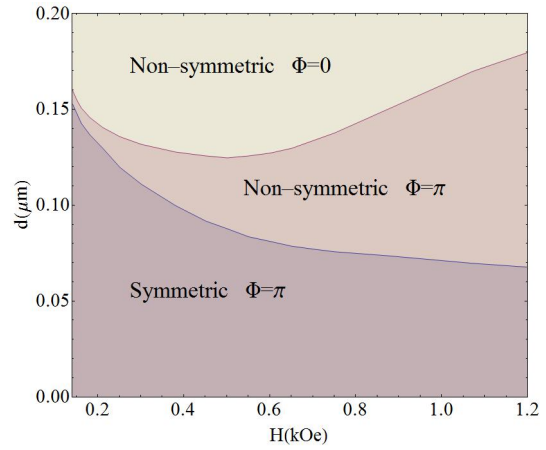

FIG. 4. The phase diagram for different values of thickness d and magnetic field H.

Zero Sound.— In two-condensate states the relative phase $\delta\phi = \phi_+ - \phi_-$ is a Goldstone mode. Its oscillation, coupled with the oscillation of the number density $\delta n = n_Q - n_{-Q}$ represents a new type of collective excitation, which we call zero sound (as in Landau's Fermi liquid, this mode is driven by the self-consistent field rather than collisions). Solving a properly modified Gross-Pitaevskii equation (see Methods), we find its spectrum. In the symmetric phase its dispersion relation is:

$$\omega = \sqrt{\frac{\hbar^2 k^4}{4m^2} + N_c \Delta \frac{k^2}{m}}. \qquad (6)$$

The magnon effective mass is of the order of the electron mass. The density of condensed magnons $n_c = N_c/V$ is about 10^{18} cm^{-3} and $\Delta \approx 10$ mK/N. The sound speed for small k in this case is $v_{0s} = \sqrt{N_c\Delta/m}$, which is about 100 m/s. Near the transition point $\Delta = 0$, the velocity of this zero sound goes to zero. For the non-symmetric case, the spectrum is:

$$\omega = \sqrt{\frac{\hbar^2 k^4}{4m^2}\kappa + N_c(B-A)(\kappa-1)\frac{k^2}{m}}, \quad (7)$$

where $\kappa \equiv \frac{(B-A)^2}{C^2}$. In the experiment of Ref. [17], $\kappa \sim 10^4$ and $B - A = 8.4$ mK/N, from which we estimate a sound speed of 3×10^3 m/s. The dispersions of zero sound for symmetric and non-symmetric cases are shown in Fig.5. Note that the range of applicability of the linear approximation decreases significantly for small C, where one of the condensate densities is small and the phase fluctuations grow.
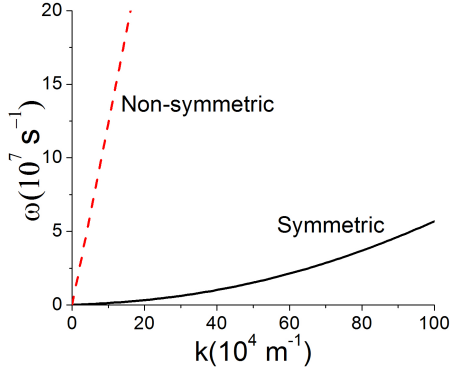

FIG. 5. Dispersion of zero sound as a function of wave vector in the direction of external magnetic field for symmetric and non-symmetric cases, respectively. For the non-symmetric case, we choose $H = 1$ kOe and $d = 5$ μm.

Domain Wall.— Since the ground state of the non-symmetric phase is doubly degenerate, it can consist of domains with different signs of δ separated by domain walls. The width w of such a domain wall is of the order of $\sqrt{\frac{\hbar^2}{2mN_c|\Delta|}}$. For the data of experiment [17] we estimate that $w \approx 10$ μm, and a domain wall energy per unit area of about 10^{-9}J/m^2.

Discussion

The ground state wave function $\Psi(z)$ generally is a superposition of two condensate amplitudes $\Psi(z) = (c_Q e^{iQz} + c_{-Q} e^{-iQz})/\sqrt{V}$, where $c_{\pm Q} = \sqrt{N_{\pm Q}}e^{i\phi_\pm}$ and V is the volume of the film. The spatial structure of $\Psi(z)$ can be measured by Brillouin Light Scattering (BLS), with intensity proportional to the condensate density $|\Psi|^2 = n_Q + n_{-Q} + 2\sqrt{n_Q n_{-Q}}\cos(2Qz + \phi_+ - \phi_-)$.

In their recent experiment, Nowik-Boltyk *el al* [17] observed the interference pattern associated with the ground state. They found that the contrast of this periodic spatial modulation is far below 100%; of the order 3%. The present theory can quantitatively explain this result. In their experiment, Ref.[17] employ $d = 5.1$ μm and $H = 1$ kOe. Then eq.(2) for A, B and C gives $A = -0.168$ mK/N, $B = 8.218$ mK/N and $C = -0.203$ mK/N, so $\Delta < 0$. This corresponds to the non-symmetric phase, where for spontaneous symmetry-breaking with $\delta = N_Q - N_{-Q} > 0$ the ratio of the numbers of magnons in the two condensates is $\frac{N_{-Q}}{N_Q} \approx \frac{C^2}{4(B-A)^2}$. The contrast is $\beta = \frac{|\Psi|^2_{max} - |\Psi|^2_{min}}{|\Psi|^2_{max} + |\Psi|^2_{min}}$. Since $C \ll B$ and $N_{-Q} \ll N_Q$, we have $\beta \approx 2\sqrt{\frac{N_{-Q}}{N_Q}} \approx \frac{|C|}{|B-A|}$. For the above values of A, B and C, β is of order 2.4%, in good agreement with experiment. The smallness of C (and A) relative to B is associated with the large parameter d/l, where $l = \sqrt{\frac{D}{\pi\gamma M}}$ is an intrinsic length scale of the system and $l \sim 10^{-6}$ m. In terms of this parameter, $\frac{|C|}{B} \sim (\frac{l}{d})^{2/3}$.

In the experiment of [17] the contrast β reaches a saturation value at a comparatively small pumping power, corresponding to the appearance of BEC. This agrees with our expression for β, which depends only on film thickness d and magnetic field H. By varying d and H, the contrast can be changed. Specifically, in the symmetric phase, $\beta = 1$; in the non-symmetric phase, $\beta < 1$; and in the completely non-symmetric case with only one condensate ($d = d^*$), $\beta = 0$. Therefore, measurement of the contrast for different values of d and H can give complete information about the phase diagram of the system, for comparison with the present theory.

Fig.6 plots C, Δ and β as functions of the film thickness d at fixed magnetic field $H = 1$ kOe. For

small d the system is in the high-contrast symmetric state. On increasing d above $d_c = 0.07$ μm, the sign of Δ changes, corresponding to the transition from the symmetric to the low-contrast non-symmetric phase. As d further increases, to $d^* = 0.17$ μm, C changes sign, and the total phase Φ changes from π to 0. Only at this point d^* does the zero-contrast phase (with only one condensate) appear. Correspondingly, a characteristic cusp in the contrast β appears near d^*.
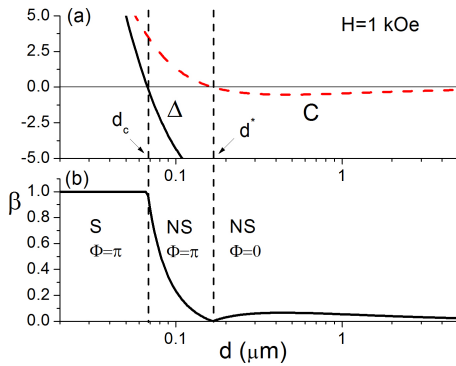

FIG. 6. (a) Phase transition criterion Δ and interaction coefficient C as functions of thickness d for fixed magnetic field $H = 1$ kOe. (b) The contrast $\beta = \frac{|\Psi|^2_{max} - |\Psi|^2_{min}}{|\Psi|^2_{max} + |\Psi|^2_{min}}$ as a function of thickness d for $H = 1$ kOe. S and NS denote symmetric and non-symmetric phase, respectively.

To conclude, we have calculated the 4-th order magnon-magnon interactions in the condensate of a film of YIG, including magnon-non-conserving terms that are responsible for the coherence of two condensates. sFor ufficiently thin YIG films ($d < 0.1$ μ) we predict a phase transition from symmetric to non-symmetric phase when the magnetic field exceeds the modest value of 0.2 kOe. We also predict that within the non-symmetric phase there is a thickness $d^*(H)$ where the modulation in the observed interference pattern should totally disappear.

Methods

Magnon Spectrum.— In a YIG film with an in-plane external magnetic field H, the magnon dispersion has been studied extensively [28–30]. At low energies, YIG can be described as a Heisenberg ferromagnet with large effective-spin $S = 14.3$ [19, 24] on a cubic lattice. The Hamiltonian consists of three parts:

$$\mathcal{H} = -J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + H_D - \gamma H \sum_i S_i^z, \quad (8)$$

the nearest neighbor exchange energy, the dipolar interaction and the Zeeman energy. We take y to be perpendicular to the film and the magnetic field to be in the plane along z. It is convenient to characterize the exchange interaction by the constant $D = 2JSa^2 = 0.24$ eV$\mathring{A}^2$. The dipolar interaction can be calculated using the method indicated in Refs.[20, 30]. The competition between the dipolar interaction and exchange interaction leads to a magnon spectrum ω_k with minima located at the two points in 2D wave-vector space given by $\mathbf{k} = (0, \pm Q)$ (i.e. along z), with an energy gap Δ_0. For film thickness $d = 5$ μm and magnetic field $H = 1$ kOe, we find that $Q = 7.5 \times 10^4$ cm^{-1} and $\Delta_0 = 2.7$ GHz. In the experiment of [17] Q was found to be about 3.5×10^4 cm^{-1}, i.e. about half the predicted value. The reason for this may be associated with a rather shallow energy minimum as a function of wavevector. In such a situation small corrections to our approximate formula can have a large effect on the value of Q. The lowest band of the magnon spectrum can be calculated using the Holstein-Primakoff transformation [32], which expresses the spin operator $\mathbf{S}$ in terms of boson operators a and $a^\dagger$.

To second order in a and $a^\dagger$, the Hamiltonian eq.(8) is:

$$\mathcal{H}_0 = \sum_{\mathbf{k}} \left[\mathcal{A}_k a_k^\dagger a_k + \frac{1}{2} \mathcal{B}_k a_k a_{-k} + \frac{1}{2} \mathcal{B}_k^* a_k^\dagger a_{-k}^\dagger \right] (9)$$

with

$$\mathcal{A}_k = \gamma H_0 + Dk^2 + \gamma 2\pi M (1 - F_k) \sin^2 \theta + \gamma 2\pi M F_k$$
$$\mathcal{B}_k = \gamma 2\pi M (1 - F_k) \sin^2 \theta - \gamma 2\pi M F_k \quad (10)$$

where $F_k \equiv (1 - e^{-kd})/kd$ and M is the magnetization of the material ($4\pi M = 1.76$ kG). Here, θ is the angle between the 2D wave vector $\mathbf{k}$ and the magnetic field direction (z). $\mathcal{H}_0$ of eq.(9) is diagonalized by the Bogoliubov transformation $a_k = u_k c_k + v_k c_{-k}^\dagger$ with $u_k = (\frac{\mathcal{A}_k + \hbar\omega_k}{2\hbar\omega_k})^{1/2}$ and $v_k = sgn(\mathcal{B}_k)(\frac{\mathcal{A}_k - \hbar\omega_k}{2\hbar\omega_k})^{1/2}$, leading to the magnon spectrum:

$$\hbar\omega_k = (\mathcal{A}_k^2 - |\mathcal{B}_k|^2)^{1/2}. \quad (11)$$

Fig.1 gives the magnon spectrum along k_z for typical values of thickness d and magnetic field H.

Number of condensed magnons $N_c = N_Q + N_{-Q}$.— Experimentally, the spin lattice relaxation time is of order 1 μs, whereas the magnon-magnon thermalization time is of order 100 ns; the magnons are long-lived enough to equilibrate before decaying, thus making BEC possible [11]. After the thermalization time the pumped magnons go to a quasi-equilibrium state with a non-zero chemical potential μ. The number of pumped magnons $N_p = N(T, \mu) - N(T, 0)$, where $N(T, \mu) = V \sum_{\mathbf{k}} \frac{1}{e^{(\omega_k - \mu)/T} - 1}$, is determined by the pumping power and the magnon lifetime. μ is a monotonically increasing function of N_p but cannot exceed the energy gap Δ_0. Therefore, on further pumping $\mu = \Delta_0$ and some of the pumped magnons fall into the condensate. The equation $N_{pc} = N(T, \Delta_0) - N(T, 0)$ thus defines the critical line for condensation. Since $\Delta_0 \ll T$ and $N_p \ll N(T, 0)$ this equation can be satisfied at a rather high temperature. The total number of condensed particles is [11, 31]

$$N_c = N_p - N(T, \Delta_0) + N(T, 0) = N(T, \mu) - N(T, \Delta_0).$$
(12)

In exactly 2D systems BEC formally does not exist since in the continuum approximation the sum in $N(T, \mu)$ diverges. However, for strong enough pumping the chemical potential approaches exponentially close to the energy gap: $\Delta_0 - \mu \approx \Delta_0 \exp(-N_p/N_0)$, where $N_0 = VTm/\hbar^2$. For $N_p/N_0 > \ln(T/\Delta_0)$ all pumped magnons occupy only one or two states $\pm Q$.

Eq.(12) determines only the total number of particles in the condensate. The distribution of the condensate particles between the two minima remains undetermined in the quadratic approximation. To resolve this issue we have shown that it is necessary to consider the fourth order terms in the Holstein-Primakoff expansion of the exchange and dipolar energy. Observe that terms of third order occur in this expansion of the dipolar interaction, but since the total momentum must be zero, such terms vanish for the condensate momenta $(0, \pm Q)$.

Zero Sound.— We now provide details about calculating the zero sound spectrum. We consider small deviations from the static symmetric solution $n_Q = n_{-Q} = n_c/2$, $\phi_+ = \pi - \phi_- = 0$, so that $n_{\pm Q} = n_c/2 + \delta n_{\pm}$ with $\delta n_+ = -\delta n_- = \delta n/2$ and $\delta\phi_+ = -\delta\phi_- = \delta\phi/2$. Then

$$E = \int dr \Big(\frac{\hbar^2}{2m} (|\nabla\Psi_+|^2 + |\nabla\Psi_-|^2)) \\ + AV(|\Psi_+|^4 + |\Psi_-|^4 + 2BV|\Psi_+|^2|\Psi_-|^2 \\ + CV(\Psi_+\Psi_- + \Psi_+^*\Psi_-^*)(|\Psi_+|^2 + |\Psi_-|^2) \Big),$$

On linearizing, the energy reads:

$$E = \int dr \Big(\frac{\hbar^2}{4mn_c} |\nabla\delta n|^2 + \frac{\hbar^2 n_c}{4m} |\nabla\delta\phi|^2 + \frac{\Delta V}{2}\delta n^2 \Big).$$

Using the commutation relation $[\delta\phi, \delta n] = i$, and the equation of motion $i\hbar\delta\dot{\phi} = [\delta\phi, H]$, we obtain:

$$\hbar\frac{\partial\delta\phi}{\partial t} = -\frac{\hbar^2}{2mn_c}\nabla^2\delta n + \Delta V\delta n, \qquad (13)$$

$$\hbar\frac{\partial\delta n}{\partial t} = \frac{\hbar^2}{2m}n_c\nabla^2\delta\phi. \qquad (14)$$

Taking Fourier transforms of the above two equations in coordinate and time, one arrives at the dispersion relations in Eq.(6).

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Additional Information

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