Stability of Bose-Einstein Condensates of Hot Magnons in Yttrium Iron Garnet Films

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We investigate the stability of the recently discovered room-temperature Bose-Einstein condensate (BEC) of magnons in yttrium iron garnet films. We show that magnon-magnon interactions depend strongly on the external field orientation, and that the BEC in current experiments is actually metastable—it only survives because of finite-size effects, and because the BEC density is very low. On the other hand a strong field applied perpendicular to the sample plane leads to a repulsive magnon-magnon interaction; we predict that a high-density room-temperature magnon BEC should then form in this perpendicular field geometry.

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In a remarkable, very recent discovery [1], a room temperature Bose-Einstein condensate (BEC) of magnon excitations was stabilized for a period of roughly 1 μs in a thin slab of the well-known insulating magnet Yttrium Iron Garnet (YIG). The pumped density of magnons was quite low ($n \sim 10^{-4}$ per lattice site), and the density n_0 of the BEC was apparently unknown, but $n_0/n \ll 1$. This result can be understood naively in terms of a weakly interacting dilute gas of bosons, provided that (i) one assumes that the number of magnons is conserved, so their chemical potential may be nonzero, and (ii) the interactions between them are repulsive (attractive interactions favor depletion of the BEC, causing a negative compressibility and instability of the BEC [2]). In the experiment, magnon-magnon collisions conserved magnon number—the decay of the BEC was attributed to spin-phonon couplings [1]. The magnons in the experiment had rather long wavelengths, $\sim\mu$m—hitherto such excitations have been treated entirely classically [3].

This experimental claim, if correct, would be the first observation of room-temperature BEC. However we show here that a rather basic question about the stability of the system needs to be answered. We find the rather startling result that in experiments reported so far, the intermagnon interactions were actually *attractive*: the BEC should have been unstable. We shall see that in the geometry used, the BEC is actually metastable to thermally activated or tunneling decay, and that it only survives because its density is very low—above a critical density, given below, it is absolutely unstable. However we then show that by changing the field configuration one can make the interactions repulsive, and the BEC should then stabilize at a much higher density—allowing for a genuine bulk room-temperature superfluid, and opening the way to much more interesting experiments.

YIG is one of the best characterized of all insulating magnets [4]. It is cubic, with lattice constant $a_0 = 12.376$ Å, ordering ferrimagnetically below $T_c = 560$ K. At room temperature the long-wavelength properties can be understood using a Hamiltonian with ferromagnetic exchange interactions between effective "block spins" $\mathbf{S}_j$, one per unit cell, whose magnitude $S_j = |\mathbf{S}_j| = a_0^3 M_s/\gamma$, with $\gamma = g_e \mu_B$, is defined by the experimental saturated magnetization density M_s ($M_s \approx 140$ G at room temperature; with $g_e \approx 2$, one has $S_j \approx 14.3$), along with dipolar couplings between these; the resulting lattice Hamiltonian takes the form

$$\hat{H} = -\gamma \sum_i \mathbf{S}_i \cdot \mathbf{H}_0 - J_0 \sum_{i,\delta} \mathbf{S}_i \mathbf{S}_{i+\delta} + U_d \sum_{i \neq j} \frac{\mathbf{S}_i \mathbf{S}_j - 3(\mathbf{S}_i \cdot \mathbf{n}_{ij})(\mathbf{S}_j \cdot \mathbf{n}_{ij})}{|r_{ij}|^3}, \quad (1)$$

where the sums i, j are taken over lattice sites at positions $\mathbf{R}_i$, etc., δ denotes nearest-neighbor spins, $\mathbf{r}_{ij} = (\mathbf{R}_i - \mathbf{R}_j)/a_0$, and $\mathbf{n}_{ij} = \mathbf{r}_{ij}/|\mathbf{r}_{ij}|$. The nearest neighbor dipolar interaction $U_d = \gamma^2/a_0^3 \approx 1.3 \times 10^{-3}$ K. The parameter J_0 is determined experimentally from $J = 2SJ_0a_0^2 \approx 0.83 \times 10^{-28}$ erg cm^2 at room temperature, implying $J_0 \approx 1.37$ K, and $U_d/J_0 \approx 0.95 \times 10^{-3}$.

In what follows we set up a theoretical description of the BEC, taking into account the external field, dipolar and exchange interactions, and boundary conditions in the finite geometry. We evaluate the interactions and the BEC stability for 2 different field configurations; the general picture then becomes clear.

(i) In-plane field: All experiments so far have had $\mathbf{H}_0$ in the slab plane. The combination of exchange, dipolar, and Zeeman couplings then leads to a magnon spectrum $\omega_{\mathbf{q}}$ shown in Fig. 1, in which the competition between dipolar and exchange interactions leads to a finite-q minimum in $\omega_{\mathbf{q}}$ at a wave vector $Q \sim 1/d$, where d is the slab thickness. To completely specify $\omega_{\mathbf{q}}$ and the intermagnon interactions one needs boundary conditions, which can involve partial pinning of the surface spins [5–7]. Demokritov *et al.* [1] assume free surface spins, implying that (i) $|(\partial M(\mathbf{r})/\partial \mathbf{r}) \cdot \mathbf{n}_s| = 0$ when $\mathbf{r}$ is at the surface; here $\mathbf{n}_s$ is the normal to the surface, and (ii) that the allowed

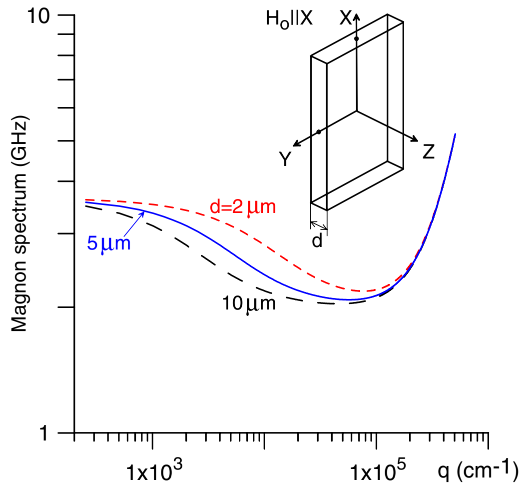

FIG. 1 (color online). The magnon spectrum, Eq. (2), for $d = 2$, 5 and 10 μm and $H_0 = 0.07$ T. The inset shows the sample geometry.

momenta along $\hat{z}$ (see Fig. 1, inset) are $q_\perp = n_\perp \pi/d$, leading to different magnon branches labeled by $n_\perp$. For now we assume a continuous in-plane momentum, and later discuss the effect of in-plane quantization, and we assume $n_\perp = 0$, taking the lowest-energy magnon branch.

The above assumptions yield an $\omega_{\mathbf{q}}$ which agrees with experiment [1], and which for $\mathbf{H}_0 \parallel \hat{x}$ takes the form [7]

$$\hbar\,\omega_q = [(\gamma H_i + Jq^2)(\gamma H_i + Jq^2 + \hbar\omega_M \mathcal{F}_q)]^{1/2}, \quad (2)$$

here $H_i = H_0 - 4\pi N_x M_s$ is the internal field, N_x the demagnetization factor (for a slab in the xy-plane, $N_x = N_y = 0$, $N_z = 1$), $\hbar\omega_M = 4\pi\gamma M_s$ (for YIG, $\hbar\omega_M \approx 0.236$ K at room temperature), and $\hbar\omega_H = \gamma H_i$. The form of the dimensionless function $\mathcal{F}_q$ depends on the direction of $\mathbf{q}$. In the important case when $\mathbf{q}$ is parallel to H_0, i.e., along $\hat{x}$, one has

$$\mathcal{F}_q \to (1 - e^{-q_x d})/(q_x d), \quad (3)$$

In the experiment [1] magnons are argued to condense at the minima $q_x = Q$; when $d = 5$ μm, $|Q| = 5.5 \times 10^6$ m^{-1}.

We now set up a theoretical description of the BEC, including all 4-magnon scattering processes (3-magnon scattering is excluded by the kinematics), using a generalized Bogoliubov quasiaverage technique [8] to describe the finite-T BEC [9]. Defining magnon operators $b_{\mathbf{q}}$, $b^\dagger_{\mathbf{q}}$, a magnon BEC at $q = \pm Q$, with n_0 condensed magnons, has quasiaverages

$$\langle b_{\pm Q}\rangle = \langle b^\dagger_{\pm Q}\rangle = \sqrt{n_0/2}, \quad (4)$$

corresponding to a condensate wave function $\Psi_Q(y) \propto \cos(Qy)$. More generally, $\Psi_Q(y)$ is multiplied by a phase factor $e^{i\phi(\mathbf{r},t)/\hbar}$, which is crucial to the BEC dynamics, but not necessary for a stability analysis of the BEC.

We make a Holstein-Primakoff magnon expansion [10] up to 4th order in magnon operators, including contributions from both (2-in-2-out) and (3-in-1-out) magnon scattering processes [11] (we ignore multiple-scattering contributions here, which are $\sim O(U_d/J_0 S) \sim 10^{-4}$ relative to the leading terms). Then, taking quasiaverages, we can write the Hamiltonian in the form

$$\mathcal{H} = \hbar\sum_q \omega_{\mathbf{q}}\left(b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + \frac{1}{2}\right) + \hat{V}^p_{\rm int} + \hat{V}^{-p}_{\rm int}, \quad (5)$$

where the interaction term $\hat{V}^p_{\rm int}$ takes the form

$$\begin{aligned}\hat{V}^p_{\rm int} = {}& n_0\left(\Gamma_0 + \frac{\Gamma_S}{4}\right)\sum_{p<Q}[b^\dagger_{Q+p}b_{Q+p} + b^\dagger_{-Q-p}b_{-Q-p}] \\ &+ \frac{\Gamma_S n_0}{4}\sum_{p<Q}[b^\dagger_{Q+p}b_{-Q+p} + b^\dagger_{-Q+p}b_{Q+p} \\ &+ b_{Q+p}b_{-Q-p} + b^\dagger_{Q+p}b^\dagger_{-Q-p}] \\ &+ \frac{\Gamma_0 n_0}{2}\sum_{p<Q}[b_{Q+p}b_{Q-p} + b_{-Q-p}b_{-Q+p} \\ &+ b^\dagger_{Q+p}b^\dagger_{Q-p} + b^\dagger_{-Q-p}b^\dagger_{-Q+p}]. \end{aligned} \quad (6)$$

Here Γ_0 and Γ_S are the four-magnon scattering amplitudes between states $(\mathbf{Q},\mathbf{Q}) \to (\mathbf{Q},\mathbf{Q})$ and $(\mathbf{Q},-\mathbf{Q}) \to (\mathbf{Q},-\mathbf{Q})$, respectively. For the sample geometry in Fig. 1, with $\mathbf{H}_0 \parallel \hat{x}$, these scattering amplitudes are found to be

$$\begin{aligned}\Gamma_0 = {}& -\frac{\hbar\omega_M}{8S}[(\alpha_1 - \alpha_3)\mathcal{F}_Q - 2\alpha_2(1 - \mathcal{F}_{2Q})] \\ &- \frac{JQ^2}{4S}[\alpha_1 - 4\alpha_2]; \end{aligned} \quad (7)$$

$$\begin{aligned}\Gamma_S = {}& \frac{\hbar\omega_M}{2S}[(\alpha_1 - \alpha_2)(1 - \mathcal{F}_{2Q}) - (\alpha_1 - \alpha_3)\mathcal{F}_Q] \\ &+ \frac{JQ^2}{S}[\alpha_1 - 2\alpha_2], \end{aligned} \quad (8)$$

with $\alpha_1 = u_Q^4 + 4u_Q^2 v_Q^2 + v_Q^4$, $\alpha_2 = 2u_Q^2 v_Q^2$ and $\alpha_3 = 3u_Q v_Q(u_Q^2 + v_Q^2)$, where $\{u_q, v_q\} = [(A_q \pm \hbar\omega_q)/2\hbar\omega_q]^{1/2}$ and A_q and B_q are given by $A_q = \gamma H_i + Jq^2 + 2\pi\gamma M_s \mathcal{F}_q$ and $B_q = -\pi\gamma M_s \mathcal{F}_q$ respectively (the magnon energy is $\hbar\omega_q = [A_q^2 - 4|B_q|^2]^{1/2}$). Higher-order multiple-scattering contributions to Γ_0, Γ_S are much smaller, $\sim O(U_d/J_0 S)$ relative to the leading terms given here [12].

We plot the combinations $2\Gamma_0 \pm \Gamma_S$ in Figs. 2 and 3. Both amplitudes are sensitive to the external field and the film thickness: $2\Gamma_0 + \Gamma_S$, becomes negative in the entire region of fields at $d < d_c \approx 2(J/\hbar\omega_M)^{1/2} \approx 0.032$ μm, whereas $2\Gamma_0 - \Gamma_S$ is positive at $d < d_c$.

Near the energy minimum (when $|p| \ll |Q|$), one has $\omega_{Q+p} \approx \omega_{Q-p}$, and the Bogoliubov transformation is straightforward because $\mathcal{H}$ is symmetric when $p \leftrightarrow -p$ and $Q \leftrightarrow -Q$. The spectrum then has 4 branches, but is

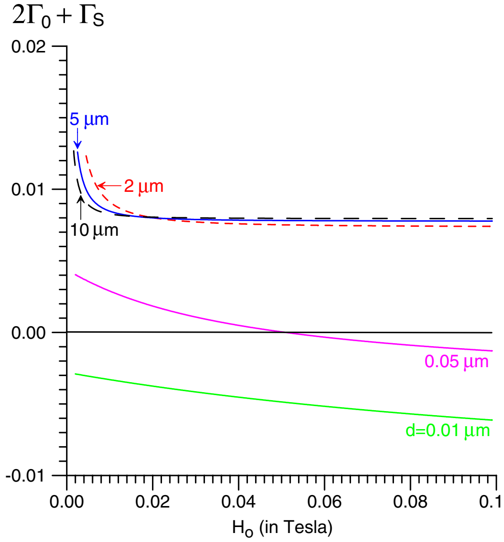

FIG. 2 (color online). The effective amplitude $2\Gamma_0 + \Gamma_S$ (in Kelvins) as a function of field H_0 at different values of the film thickness d. Dashed lines: $d = 2$ and 10 μm. Solid lines: $d =$ 0.01, 0.05, and 5 μm.

twofold degenerate, with quasiparticle energies

$$\epsilon_{p\eta} = \sqrt{\Omega_Q(p)[\Omega_Q(p) + n_0(2\Gamma_0 + \eta\Gamma_S)]}, \qquad (9)$$

where $\eta = \pm 1$, and $\Omega_Q(p) = \hbar(\omega_{Q+p} - \omega_Q)$.

The inset in Fig. 3 shows the "phase diagram" of the quasi-two-dimensional YIG for different values of d and H_0^x. One immediately sees a paradox: the amplitudes are never both positive, so bulk BEC should not exist—yet BEC has been observed [1] in samples with an in-plane field $H_0^x \sim 0.07$ T, in which $d \sim 5$ μm.

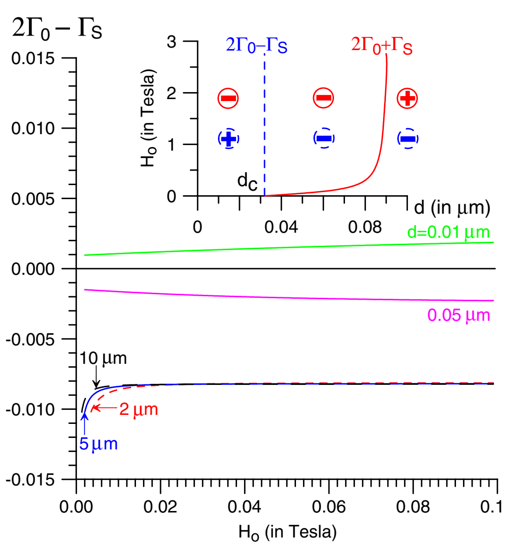

FIG. 3 (color online). The effective amplitude $2\Gamma_0 - \Gamma_S$ (in Kelvins) as a function of external field H_0 at different values of the film thickness d. Dashed lines: $d = 2$ and 10 μm. Solid line: $d = 0.01, 0.05$, and 5 μm. The inset shows the "phase diagram" of YIG in the (H_0, d) plane. The amplitude $2\Gamma_0 + \Gamma_S$ is positive everywhere to the right of the solid line (the region ⊕). The amplitude $2\Gamma_0 - \Gamma_S$ is negative everywhere to the right of the dashed line (the region ⊖).

The paradox is resolved by noting that in a finite geometry, the energy gap from the condensate to excited modes can be larger than the scattering amplitude, leading to a potential barrier to decay. For weakly interacting Bose gases this yields [13] an upper critical number $n_0^{\rm cr}$ of condensate particles, above which the barrier disappears; one has $\alpha_0 n_0^{\rm cr}/l_0 = k$, where α_0 is the s-wave scattering length, and l_0 the characteristic size of the BEC wave function. The constant $k \sim O(1)$, and depends on the sample geometry.

In the present case we can write the critical density $n_0^{\rm cr}$ as $|2\Gamma_0 - \Gamma_S| n_0^{\rm cr} \sim \epsilon_{\mathbf{p}}^{\rm min}$, where $\epsilon_{\mathbf{p}}^{\rm min}$ is the minimum quasiparticle energy in the presence of the BEC; below $n_0^{\rm cr}$ the BEC is metastable to tunneling or thermal activation. If the BEC were to spread through the entire slab, then $n_0^{\rm cr}|2\Gamma_0 - \Gamma_S| = \Omega_Q(\pi/L)$, where the length L depends on the direction of the field relative to the slab axes. Taking this result literally for the experiment [1], with a slab measuring 20×2 mm^2 in the plane, one finds $10^{-8} < n_0^{\rm cr} < 10^{-6}$ (for fields along the long and short sides of the slab, respectively). However this result is certainly too low, since it assumes a perfectly uniform BEC—in reality disorder and edge effects will smear the magnon spectrum and restrict the size of the BEC. A more realistic estimate for L is then $L_{\rm eff} = (L_x L_y L_z)^{1/3} \sim 0.6$ mm, giving $n_0^{\rm cr} \sim 10^{-5}$ for the experiment. Thus we conclude that for in-plane fields, it will be impossible to raise n_0 above this value; this could be checked experimentally (e.g., by increasing the pumping rate).

*(ii) Perpendicular field:*Now the results are very different. Consider first an infinite thin slab, which is simple to analyze. The competition between the external field $\mathbf{H}_0 = \hat{z}H_0$ and the demagnetization field (which favors in-plane magnetization) gradually pulls the spins out of the plane; below a critical field $H_c = \hbar\omega_M/\gamma \approx 1760$ G, the Free Energy is degenerate with respect to rotation around $\hat{z}$ and so the magnons are gapless, but at H_c they align with $\mathbf{H}_0$ and a gap $\hbar\omega_0 \equiv \hbar\omega_{q=0} = \gamma H_0 - \hbar\omega_M$ opens up. The minimum in the magnon spectrum is always at $q = 0$ (Fig. 4).

The intermagnon scattering amplitude Γ is now always positive

$$\Gamma(q = 0) = \frac{\hbar\omega_M}{4S}. \qquad (10)$$

This radically changes the situation—now a BEC is stable with no restriction on the condensate density. There are however restrictions on H_0; when 2200 G $< H_0^z <$ 3500 G the system has a "kinetic instability" [14], in which the pumping of the magnons at one frequency destabilizes the magnon distribution, along with strong microwave emission.

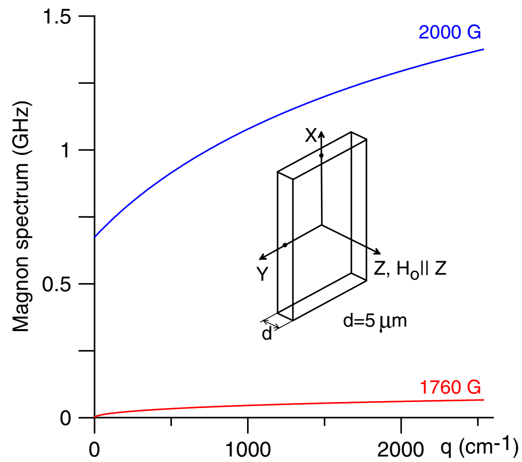

FIG. 4 (color online). The magnon spectrum for an infinite slab of YIG, assuming free surface spin boundary conditions, with magnetization polarized perpendicular to the plane (see inset). The spectra are shown for $H_0^z = H_c \approx 1760$ G where the spectrum is gapless, and for $H_0^z = 2000$ G.

In a real finite sample things are much more complicated. Even without surface anisotropy the spins near the slab edge are put out of alignment with the bulk spins by edge demagnetization fields, and surface anisotropy does the same to spins on the slab faces. However in the central region of the sample, at distances further from the surface than the exchange length, the spectrum returns to the infinite plane form. For fields well above H_c, e.g., for $H_0^z \sim 2000$ G, all of the spins will be aligned along $\hat{z}$, and (10) will then be valid everywhere.

In this case we have the striking result that a BEC of pumped magnons should be possible with densities much higher than present. To give an upper bound is complicated since the problem then becomes essentially nonperturbative (similar to, e.g., liquid ^{4}He), beyond the range of higher-order magnon expansions. However, there appears to be no obstacle in principle to raising $n_0/n \sim O(1)$. At present the highest achievable density is probably limited by experimental pumping strengths rather than any fundamental restrictions. Such a high-density BEC existing at room temperature would be extremely interesting, and certainly possess unusual magnetic properties.

*Remarks:*Our 2 main results are: (i) current experiments have all been done in a regime where the BEC is absolutely unstable (and only survives metastably at very low density because of the finite geometry); nevertheless (ii) we show that experiments done in a transverse field should see high-density bulk BEC in YIG slabs. The 2 cases studied above are actually limiting cases of a more general situation in which one can manipulate the intermagnon interactions by varying the field direction and strength, and vary the upper critical density for BEC formation by changing the sample geometry. Thus the analysis can be easily generalized to long "magnetic wires" or whiskers, and we also expect that BEC will be stabilized there when the external field is perpendicular to the sample axis, but unstable or metastable when the field is parallel to the sample axis. Further details of the various possible cases will be published elsewhere.

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