Quantum Paradigm of the Foldover Magnetic Resonance.

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The explosive development of quantum magnonics requires considering several previously known effects from a new angle. In this article, we revise the phenomenon of "foldover" (bi-stable) magnetic resonance from the point of view of quantum magnonics. The density of magnons under strong excitation can exceed the critical value for the formation of a magnon Bose condensate. Under these conditions, the effect of quantum transport of magnons should be considered. In particular, the effect of spin superfluidity, discovered earlier in superfluid \textsuperscript{3}He should lead to spatial redistribution of the precessing magnetization. Our experimental results confirm a significant change in properties of the foldover magnetic resonance in yttrium iron garnet (YIG) due to superfluid magnetization transport. This discovery paves the way for many quantum applications of supermagnonics, such as magnetic Josephson effect, long-distance spin transport, Q-bit, quantum logics, magnetic sensors, and others.

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INTRODUCTION

The purpose of our investigations is a modern analysis of the so-called folded magnetic resonance taking into account the results of breakthrough discoveries of the coherent transport properties of magnons at its high concentration. Usually, the dynamic properties of the deflected magnetization are described in terms of the Landau-Lifshitz-Gilbert phenomenological model. This approach uses classical mechanics and correctly describes magnetic systems at low levels of excitation. According to the Holstein - Primakoff transformation [1], a magnetically ordered system consists of a ground state (which plays the role of quantum vacuum) and a gas of quantum excitations - magnons, which obey Bose statistics. The density of magnons can change due to their creation and annihilation from the vacuum. At a finite temperature, the concentration of thermally activated magnons is low, and the spin dynamics can be described in the classical approximation. However, the concentration of magnons can be significantly increased by introducing non-equilibrium magnons. According to Bose statistics, magnons should form a Bose-Einstein condensate (mBEC) at a high concentration and obey the laws of quantum physics. In this case, quantum transport phenomena such as superfluidity should take place.

The first mBEC state was experimentally discovered in the antiferromagnetic superfluid \textsuperscript{3}He-B as a state emitting a very long-lived induction signal even in a strongly inhomogeneous magnetic field [2, 3]. In these experiments, an exciting radio frequency (RF) pulse produced a high density of magnons, which emit a typical induction signal. This signal decays due to the inhomogeneity of the magnetic field. However, with a certain time delay, the coherence of magnons recovered spontaneously and emits a very long coherent signal. A topologically protected mBEC emitting minute-long signals were discovered and explained in [4, 5]. This signal can be thought of as a time crystal [6]. It was found that mBEC can be maintained permanently when the relaxation (evaporation) of magnons is compensated by the creation of new magnons from a vacuum using high frequency pumping [7, 8].

In the case of repulsive interaction between non-equilibrium magnons, the mBEC state can exhibit the properties of quantum spin transport - spin superfluidity. The positive interaction energy is required to compensate for the kinetic energy of the magnon superflow. The balance between these energies determines the critical Landau flow of magnons and the critical gradient of phase $\nabla \alpha_c$, as well as the Ginzburg-Landau coherence length $\xi_{GL}$:

$$\nabla \alpha_c = 1/\xi_{GL} = \sqrt{\omega_0 \Delta \omega}/c_{SW},$$

(1)

where $c_{SW}$ is a spin wave velocity and $\Delta \omega$ is a frequency shift due to magnons interaction. The long distance spin supercurrent, Landau critical velocity, and phase slippages were observed in the experiments, described in [7, 10, 11]. Other quantum phenomena have also been observed, such as the magnonic Josephson phenomena [12, 13] and magnon quantum vortices [14, 15]. In particular, it is very important for this article that typical foldover magnetic resonance signals are observed in these experiments [7, 8]. The properties of these signals have been well understood quantitatively and have been used to study the quantum properties of magnonic BECs in superfluid \textsuperscript{3}He-B [16–18]. The comprehensive review of...
these phenomena can be found in [19, 20].

QUANTUM PHENOMENA IN SOLID MAGNETS

Observation of quantum spin phenomena in superfluid $^3$He-3 is quite expected since it is a quantum liquid. However, the question arose whether such quantum phenomena could be observed in solid magnetic materials. Essentially, magnetism is a quantum phenomenon because spin is of quantum origin. However, for many cases, magnetic phenomena can be described in the classical approximation, when a large ensemble of spins is described by phenomenological theories that take into account only the average value of the local magnetization. The phenomenon of spin transfer in solid magnets is sometimes considered by analogy with superfluid phenomena [21, 22]. Despite the fact that its mathematical description has a formal similarity to superfluidity, it should be borne in mind that superfluidity and Bose condensation are purely quantum phenomena. Therefore, it makes no sense to consider them within the framework of classical models, which deal with the spin waves and magnetization precession.

For description of magnetic quantum phenomena the paradigm of the Holstein-Primakoff transformation should be applied [1]. Magnon quantum phenomena occur at a sufficiently high concentration of magnons at the conditions of Bose-Einstein condensate formation. Strictly speaking, the properties of mBECs are beyond the scope of classical physics and are traditionally described by the Gross-Pitaevskii formalism developed to describe the atomic Bose condensate [23]. The magnon density required for the formation of the Bose condensate is easy to calculate for various magnetically ordered substances, as shown in [24].

THEORETICAL BACKGROUND

A magnetically ordered state can be represented as a stationary ground state with a gas of thermal excitations - magnons, described by Bose statistics, due to the fact that magnons have a spin equal to 1:

\[ n_k = \left( \exp \frac{\epsilon_k - \mu}{k_B T} - 1 \right)^{-1}. \]  

(2)

It is easy to see an analogy with the Universe, where elementary particles are excitations of a quantum vacuum. In contrast, magnons are quasiparticles, the density of which is in thermal equilibrium with the magnetic system. But we can increase this density by pumping new, non-equilibrium magnons using magnetic resonance. In the "classical" consideration, this is described as a deflection of the magnetization $\hat{M}$ by an angle $\beta$. In the language of magnons, this process excites an additional magnons, the number of which can be estimated as:

\[ \hat{N} = \hat{a}^\dagger \hat{a} = \frac{\mathcal{S} - \hat{S}_z}{\hbar}, \]  

(3)

where $\hat{a}^\dagger$ and $\hat{a}$ are operators of magnon annihilation and creation. This equation relates the number of excited magnons $\hat{N}$ to the deviation of spin $\hat{S}_z$ from its equilibrium value

\[ \hat{S}_z^0 = \mathcal{S} = \frac{\chi H}{\gamma} = \hat{M}, \]  

(4)

where $\chi$ is the susceptibility and $\gamma$ is the gyromagnetic ratio. The density of magnons can be so high that mBEC state forms. Magnon BEC is a quantum phenomenon and has the same off-diagonal long-range order (ODLRO) signature as a conventional superfluid:

\[ S_x + i S_y = \langle \hat{S}^+ \rangle = S \sin \beta e^{i(\alpha + \omega t)}. \]  

(5)

Here $\hat{S}^+$ is the operator of creation of spin in $z$ direction; $\beta$ is the tipping angle of precessing magnetization; $\omega$ is the global frequency of precession; and $\alpha$ is the phase of precession, which is coherent over the sample. The corresponding wave function has a form:

\[ \Psi = \sqrt{\frac{2S}{\hbar}} \sin \frac{\beta}{2} e^{i(\alpha + \omega t)}. \]  

(6)

This means that the coherent precession of the magnetization can be regarded as a Bose condensation of magnons. The role of the magnon chemical potential, $\mu_M = dE/dN_M$, is played by the frequency of precession $\omega = dE/dS_z$. This frequency is global - i.e. it is constant across the whole sample - in the same way as the chemical potential in the thermodynamic system.

There are two approaches to study the thermodynamics of atomic systems: at fixed particle number $N$ or at fixed chemical potential $\mu$. For the magnon BEC, these two approaches correspond to two different experimental arrangements: the pulsed and continuous one. In the case of free precession after the pulse, the number of magnons pumped into the system is conserved (if one neglects the losses of spin). This corresponds to the situation with the fixed $N_M$, in which the system itself will choose the global frequency of the coherent precession - the magnon chemical potential, $\omega = \mu_M$. The opposite case corresponds to a continuous wave resonance, when a small RF field is applied to compensate the relaxation. In this case the frequency of precession is fixed by the frequency of the RF field,

\[ \mu_M \equiv \omega = \omega_{RF}, \]  

(7)

and now the number of magnons $N_M$ will be adjusted to this frequency to match the resonance condition. This
In the case of a spatial inhomogeneity of the magnetic field, a spatial gradient of the precession phase $\alpha$ arises, which is directly related to the phase of the wave function of magnons and leads to the magnon superfluid current [16]:

$$\mathbf{J} = N \nabla \alpha,$$  

which redistributes magnons until the field inhomogeneity is compensated by frequency shift. And this is possible only in the case of positive frequency shift with the magnons density, i.e. for the repulsive interaction between magnons. This process is in many ways analogous to the removal of the magnetic field from superconductors (Meissner effect) and leads to the formation of a region with coherent precession of magnetization. The essence of this process is described in detail in the articles [25, 26].

After a superfluid $^3$He the mBEC and spin superflow were observed in antiferromagnets with coupled nuclear-electron precession CsMnF$_3$ [27] and MnCO$_3$ [28, 29], where the positive frequency shift from the magnon density takes place.

In this article, we are considering the experimental properties of mBEC in out of plane magnetized YIG films. The precession frequency depends on the angle of magnetization deflection $\beta$ [30]:

$$\omega_N = \omega_0 - \gamma 4\pi M_S \cos \beta,$$

where $\omega_0 - \gamma 4\pi M_S$ is the frequency at low excitation, which is determined by the external field (the first term) and the demagnetizing field (second term). The positive frequency shift is also clearly seen. The frequency increases with the deviation of magnetization, that is, with an increase in the number of non-equilibrium magnons ($1 - \cos \beta$). Therefore the uniform precession of the magnetization is stable in this configuration. [31]. It was shown in the work [24] that the critical magnon density in this configuration is achieved with a deviation of the magnetization by only about $3^\circ$. At high excitation, the quantum properties of mBEC should be taken into account.

When studying YIG films under strong excitation conditions, the effect of nonlinear resonance was found, in which the precession frequency depends on the amplitude of its excitation (foldover resonance) [32]. An approximate analytical solution of the Landau-Lifshitz equations under nonlinear resonance conditions can be obtained only for a single oscillator model in the limit of a relatively small frequency shift [33]. The effective magnetic field in macroscopic samples is inhomogeneous due to edge effects, spatial inhomogeneity, and some defects. Therefore, the resonance must be described by a set of coupled oscillators. The theoretical analysis is complicated by the fact that the excitation of the resonance is also spatially inhomogeneous, especially when it is excited by a strip line. In addition to the local Gilbert damping, one should also take into account the relaxation processes associated with spin diffusion in the case of spatial inhomogeneity of the resonance, as well as the interaction with the metallic strip line [33]. All these circumstances lead to the inability to construct a quantitative theory describing the real signals of foldover resonance in YIG films [34]. The use of microsimulation programs is also limited, as the time, required for calculations, increases dramatically with increasing sample size. However, the quantum properties of magnons can be used to describe the magnons behavior in the case of nonlinear resonance in YIG films at the conditions of mBEC. And the main result of this approach is that the state of the system is determined not by the amplitude of excitation, but also by its frequency, as follows from the Eq. (7).

THE EXPERIMENAL RESULTS

We investigated the ferromagnetic resonance adsorption signals in the YIG film at a frequency of 9.26 GHz and at different RF excitation powers. The experiments were performed on YIG films of 6 and 1 $\mu$m thicknesses in a shape of a disks 0.5 and 0.3 mm in diameter. For a detailed description of the experiments, see the Methods section. The signals from first sample are shown in Fig. 1. At first sight it looks like a well known signals of the non-linear foldover resonance, first described in [32]. Indeed this theory does not well corresponds to the experimental results at a relatively high excitation [34]. We have found the reason of Anderson model failer. It didn’t take in to account the mBEC and spin supercurrents, which play a very important role at the high angles of magnetization deflection.

First of all let us pay attention to signals at small excitation powers of 0.05, 0.1 and 0.4 mW (see inset in Fig. 1). The amplitude of adsorption signal grows proportionally to the RF field. It corresponds to a linear excitation of magnon gas. At higher excitation the “capture” of a signal takes place, as shown in Fig. 1. The signal begins to follow the sweeping field. At a field shift of more than 2.5 Oe, magnon quantum effects must be taken into account, since this shift corresponds to angles of magnetization deflection more than $3^\circ$. At this angle, the magnon density corresponds to the formation of the magnon BEC [24]. At a 80 mW excitation the signal follows the magnetic field shift up to 80 Oe, which corresponds to a frequency shift of about 220 MHz and angle of magnetization deflection about $18^\circ$. The observed signals are very robust and stable. They have the similar properties for the different samples, we have investigated. At some critical field shift the signal fails over (the FO point). The position of this point strongly depends on
FIG. 1: Amplitude of the absorption signal at different RF pump power in a decreasing magnetic field. The enlarged scale is shown in the inset. Here and in the next figures signals marked a - h corresponds to an RF power 80, 40, 20, 10, 1, 0.4, 0.1 and 0.05 mW.

FIG. 2: The energy dissipated by a magnon spin system at different level of exciting power. The energy was calculated as a product of absorption signal on the amplitude of magnetic field. The enlarged scale is shown in the inset.

The reason for the signal destruction is that the RF pump energy becomes insufficient to maintain a coherent state. The energy, absorbed by the mBEC state, is determined by the processes of magnons relaxation. Its equivalence means that the magnon state depends only on the shift and not from the pumping energy. This is the property of mBEC at a continuous pumping, described by the Eq. (7). In the precession frame, where both RF fields and deflected magnetization $M_\perp$ are constant, the interaction energy term is

$$F_{RF} = -H_{RF} M_\perp \cos(\alpha - \alpha_{RF}),$$

(10)

where $\alpha_{RF}$ is the phase of RF field. This term softly breaks the $U(1)$-symmetry and serves as a source of the mass of Nambu-Goldstone mode of mBEC [35].

The phase difference between the condensate and RF field $(\alpha - \alpha_{RF})$ is determined by the energy losses due to magnetic relaxation, which is compensated by the pumping power of the RF field:

$$W = \omega M_\perp H_{RF} \sin \beta \sin (\alpha - \alpha_{RF}).$$

(11)

The phase difference is automatically adjusted to compensate the losses. If dissipation is small, the phase shift is small $(\alpha - \alpha_{RF} \ll 1)$ and can be neglected. The neglected $(\alpha - \alpha_{RF})^2$ term leads to the nonzero mass of the Goldstone boson – quantum of the second sound waves on mBEC state in the magnonic superfluid [35]. The signal breaks down at the moment, when the RF power is not enough to compensate the magnons dissipation. Since the pumping (11) is proportional to $\sin \beta \sin(\alpha - \alpha_{RF})$, a critical tilting angle $\beta_c$, at which the pumping cannot compensate the losses, increases with increasing $H_{RF}$. The breaks down should occurs when the phase shift $(\alpha - \alpha_{RF})$ reaches $90^\circ$. However, due to the inhomogeneity of relaxation, it can appear at a smaller average angle [8]. The main process of magnetic relaxation in the
iron garnet film is determined by Gilbert dumping, which is proportional to the deflection angle of the precessing magnetization $\beta$ [36]. Usually the coefficient of Gilbert damping $\alpha_G$ is estimated from the homogeneous broadening of resonance line $\delta H / H$. For our sample it is about $2 \times 10^{-4}$ as follows from Fig. 1. However, the method for estimating $\alpha_G$ from line broadening is not reliable, since it can be mixed with inhomogeneous line broadening. From the result of our experiments we can suggest a new method of $\alpha_G$ estimation which is not sensitive to inhomogeneous broadening. The energy, dissipated by mBEC due to damping is equal to

$$W_\perp = \sigma \alpha_G^2 M^2 S \sin^2 \beta,$$

(12)

where $\sigma$ is a spectrometer parameter that relates the pump power, the amplitude of the RF field, and the ratio of the cavity and sample volumes. The dissipated energy as a function of the magnetization deflection, recalculated from the field shift is shown in Fig. 3. The perfect square dependence confirms that the main source of dissipation is the Gilbert damping. The estimations of spectrometer parameter $\sigma$ correspond well to $\alpha_G$ of about $2 \times 10^{-4}$.

The foldover resonance is usually characterized by a big difference between the fields of signal dropout at a field sweep down $(H_{dn})$ and its recovering at a sweep field up $(H_{up})$ [33]. This follows from the property of a single non-linear oscillator, shown in inset in Fig. 5. The $H_{up}$ field corresponds to the end point of the second branch of the solution and is located relatively close to the linear resonance field. This model describes well the results of experiments with a nanosized YIG sample [37]. But this does not apply to macroscopic samples. In our case the difference between $(H_{dn})$ and $(H_{up})$ is much smaller as shown in Fig. 4 by the filled symbols. This difference can be explained by a non-resonance excitation, well known for mBEC in 3He-B [20] and in solid-state antiferromagnets [38]. The similar effect was observed also in the in-plane magnetized YIG film [39]. It consists of the fact that the RF field excites spin oscillation modes with energies higher than homogeneous resonance. As a result, the magnon density increases and a magnonic BEC is formed. This process is shown in Fig. 5. When the field is swept up, resonance first appears at the edge where the local field is higher. The spin supercurrent distributes magnons in the central part of the sample, and the global mBEC is restored. The distribution of mBEC from the excitation region to the lower magnetic field in a YIG film placed in a magnetic field gradient has recently been investigated and confirmed in work [40].

FIG. 4: The dependence of fields $H_{dn}$ and $H_{up}$ in units of field shift devied by the line broadening $(\Delta H_0)$ as a function of effective RF field. The open symbols shows the theoretical curve for a single non-linear oscillator, and filled symbols - our experimental data. The field $H_{dn}$ is adjusted to the theoretical value for a nonlinear oscillator. Inset: the solution for non-linear oscillator (dashed line) in comparison with and experimental data (dotted blue line). The difference in the $H_{up}$ field is clearly seen.

FIG. 5: Profile of the local effective magnetic field due to edge of the sample (red line) calculated by a microspin program. The angle of magnetization deflection in mBEC (shown by dotted line) is distributed in the way that the frequency shift-compensate the inhomogeneity of magnetic field and all magnetization is precessing in the frequency $\omega_{RF}$ (dashed line). In inset the process of signal restoration at field sweep up is shown. The resonance appears first at the edge. The spin supercurrent distribute magnons to a central part and the signal of foldover resonance restored.

Conclusion

Our experimental results confirm a significant change in the properties of folded magnetic resonance in YIG due to the quantum properties of mBEC and the formation of a state with coherent precession. This state can be thought of as a quantum object that can be used for quantum calculations. Entanglement between two BEC states was suggested by [41], and the Josephson effect be-
between two BECs was demonstrated in a superfluid $^3$He-B recently [42]. The important point is that the mBEC qubit can be used even at room temperature. This state is the first permanent superfluid state of condensed matter at room temperature. It is an ideal platform for development of microwave magnetic technologies, which have already resulted in the creation of the magnon transistor and the first magnon logic gate [43, 44]. The YIG can be used as the basis for new solid-state quantum measurement and information processing technologies including cavity-based QED, optomagnonics, and optomechanics [45]. This discovery paves a way to many quantum applications, such as magnetic Josephson effect, long distance spin transport, Q-bit, quantum logics, magnetic sensors and others. It can be considered as a new branch of modern magnetism - supermagnonics.

Methods

All samples were prepared from the yttrium iron garnet films grown by liquid phase epitaxy on 500 $\mu$m thick GGG substrates with the (111) crystallographic orientation [46]. To reduce the effect of cubic magnetic anisotropy, we used scandium — substituted $Lu_{1.5}Y_{1.5}Fe_{4.4}Sc_{0.6}O_{12}$ iron garnet films; the introduction of lutetium ions was necessary to match the parameters of the substrate and film crystal gratings. It is known that the introduction of scandium ions in such an amount reduces the field of cubic anisotropy by more than an order of magnitude [47]. In addition, the used lutetium and scandium ions practically do not contribute to additional relaxation in the YIG. The samples were prepared in the form of the disks with diameters of 500 and 300 $\mu$m and a thickness of 6 $\mu$m. The disks were made by photolithography. To avoid magnetic pinning on the surface the sample was etched in a hot phosphoric acid [48]. As a result, the edges of the disk have a slope of 45 degrees and had a smooth surface.

The CW ferromagnetic resonance experiments were performed on Varian E-12 X-band EPR spectrometer at the room temperature and the frequency 9.26 GHz. The RF field was oriented in plane of the samples. The amplitude and the frequency of magnetic field modulation were 0.05 Oe and 100 kHz, respectively. This frequency is much lower than the estimated frequency of the second sound of the magnon BEC (The Goldston mode). That is why we may consider these conditions as stationary. The absorption signals, presented here, were obtained after the integration of the original signals.

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