Nonequilibrium Bose-Einstein condensation of hot magnons

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We present an analysis of the emergence of a nonequilibrium Bose-Einstein-type condensation of magnons in radio-frequency pumped magnetic thin films, which has recently been experimentally observed. A complete description of all the nonequilibrium processes involved is given. It is demonstrated that the phenomenon is another example of the emergence of Bose-Einstein-type condensation in nonequilibrium many-boson systems embedded in a thermal bath, a phenomenon evidenced decades ago by the renowned late Herbert Fröhlich.

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Recently have been reported several experimental studies on the kinetic of evolution of the system of spins in thin films of yttrium-iron garnets (YIGs) in the presence of a constant magnetic field and being excited by a source of radio-frequency (rf) radiation which drives the system out of equilibrium.\(^1\)\(^2\) The experimental results evidence the emergence of what has been dubbed as a nonequilibrium Bose-Einstein condensation (BEC), consisting in a large enhancement of the population of the magnons in the state lowest in energy in their energy-dispersion relation. As we show here, it is another example of a phenomenon common to many-boson systems embedded in a thermal bath (in the conditions that the interaction of both generates nonlinear processes) when driven sufficiently away from equilibrium by the action of an external pumping source and which display possible applications in the technologies of devices and medical applications. One case was evidenced by Herbert Fröhlich who considered the many-boson system consisting of polar vibration (LO phonons) in biopolymers under dark excitation (metabolic energy pumping) and embedded in a surrounding fluid.\(^3\)\(^6\) Excitons\(^7\)\(^8\) and acoustic phonons\(^9\)\(^11\) are other studied cases.

In the present Rapid communication, we present a full analysis of the kinetic evolution of hot magnons. This is done by means of the characterization of the macroscopic state of the magnon system in terms of the thermomechanical statistics based on the framework of a nonequilibrium statistical ensemble formalism (NESEF for short).\(^12\)\(^16\) Other tentatives, along different nonthermostatistic approaches, has been provided, see, for example, Refs. 17–19. The one by Tupitsyn et al.\(^17\) considers that the intermagnon interaction in a YIG thin film magnetized in the plane prevent the conditions for stabilization in the BEC. This is shown to be nonvalid in Ref. 18, where Rezende explains the generation of quantum coherence in a macroscopic state, created by a proposed coupling with a thermal bath of hot magnons. According to our results, based on the fundamental works of Herbert Fröhlich, the phenomenon is a general result generated by Fröhlich effect. The work of Malomed et al.\(^19\) refers to a propagation of solitons (the macroscopic wave function of the coherent states), which also arise out of Fröhlich effect and shall be reported in future publication (for other cases of boson quasiparticles see Refs. 10 and 8).

Let us consider a system of \(N\) localized spins in the presence of a constant magnetic field, being pumped by a rf source of radiation driving them out of equilibrium while embedded in a thermal bath consisting of the phonon system (the lattice vibrations) in equilibrium with an external reservoir at temperature \(T\). The Hamiltonian of the system of magnons and phonons is derived from the full Hamiltonian of spins and lattice vibrations after going through Holstein-Primakoff and Bogoliubov transformations.\(^20\)\(^21\) It consists of the Hamiltonian of the free magnons and phonons, \(\hat{H}_{\text{SS}}\) and \(\hat{H}_{\text{SL}}\), the magnon-magnon interaction originated on the spins exchange and dipolar interactions, \(\hat{H}_{\text{SS}}\), the magnon-phonon interaction, \(\hat{H}_{\text{SL}}\) (from spin-lattice interaction), spin-orbit interaction, and the interaction with the external rf-field radiation, and in the presence of the sample black-body radiation, all having the usual known forms.\(^20\)\(^21\)

In terms of the dynamics generated by this full Hamiltonian the equations of evolution of the macroscopic state of the system are obtained. As already noticed we resort to the statistical thermomechanical formalism NESEF.\(^12\)\(^16\) The formalism introduces the fundamental properties of historicity and irreversibility in the evolution of the nonequilibrium system where dissipative and pumping processes are underway. First step in the application of the formalism consists in the choice of a basic set of variables that should characterize the macroscopic state of the system (the appropriate nonequilibrium thermodynamic state of the system).\(^22\)\(^23\) At the macroscopic level, for the phonons is taken the Hamiltonian \(\hat{H}_{\text{SL}}\) and for the magnon system is taken the single-magnon reduced density matrix (Wigner-von Neumann density operator)\(^24\)\(^28\) written in second quantization, and, being bosons, are also introduced the amplitudes, i.e., the creation, \(c_{\mathbf{q}}^\dagger\) and annihilation, \(c_{\mathbf{q}}\), operators in magnon states whose eigenstates are the so-called coherent states. Hence, the set of basic microscopic variables is then composed of

\[
\{ \hat{N}_{\mathbf{q}} = c_{\mathbf{q}}^\dagger c_{\mathbf{q}} , c_{\mathbf{q}} \} \hat{H}_{\text{SL}} \}
\]

with \(\mathbf{q}\) running over the Brillouin zone. The single-magnon density matrix is composed of the diagonal elements \(\hat{N}_{\mathbf{q}} = c_{\mathbf{q}}^\dagger c_{\mathbf{q}}\) (called populations) and the nondiagonal ones \(\hat{N}_{\mathbf{q},\mathbf{Q}} = c_{\mathbf{q}+\mathbf{Q}}^\dagger c_{\mathbf{q}} c_{\mathbf{q}} - \hat{N}_{\mathbf{q},\mathbf{Q}}^\dagger c_{\mathbf{q}} c_{\mathbf{q}}\) with \(\mathbf{Q} \neq 0\). The latter, describing the local inhomogeneities of the populations \(\hat{N}_{\mathbf{q}}\), are not relevant for the present problem once space-resolved experiments are
not involved. It can be noticed that all the single-particle observables of the system are expressed in terms of the single-particle reduced density matrix.\textsuperscript{25} Finally, the space of nonequilibrium thermodynamic variables consists of the average values over the nonequilibrium ensemble of the quantities in set, Eq. (1), say,

\begin{equation}
\{N_q(t), \langle c_q(t) \rangle, \langle c_q^\dagger(t) \rangle, E_i \}.
\end{equation}

The equations of evolution for these variables are the quantum-mechanical equations of motion for the dynamical quantities of set, Eq. (1), averaged over the nonequilibrium ensemble. They are handled resorting to the NESEF-based nonlinear quantum kinetic theory,\textsuperscript{12,15,27} with the calculations performed in the approximation that incorporates only terms quadratic in the interaction strength—with memory and vertex renormalizations neglected.\textsuperscript{27,28} The resulting evolution equations acquire quite cumbersome expressions, which here, for just discussing the physics involved and the origin of the phenomena, are not explicitly written down. We present them in a compact form, indicating and describing the contribution of the different processes involved, which for the populations are

\begin{equation}
\frac{d}{dt} N_q(t) = S_q(t) + L_q(t) + L_q(t) + F_q(t) + M_q(t) + A_q(t),
\end{equation}

where on the right we do have: (i) \( S_q(t) \) is the rate of growth of the population in \( q \) mode produced by the external source, which is composed of three contributions, namely, a direct production, a positive feedback (only associated to parallel pumping excitation), and a term of decay through photon emission, the latter leading to saturation of absorption when under continuous excitation; (ii) \( L_q(t) \) is a term of linear relaxation to the lattice with a relaxation time \( \tau_q^{\text{relax}} \); (iii) \( L_q(t) \) is a term involving nonlinear relaxation to the lattice, referred to as the Livshits contribution;\textsuperscript{29} (iv) \( F_q(t) \) is a peculiar and fundamental contribution of a nonlinear character arising out of the magnon-lattice interaction, which takes the form

\begin{equation}
F_q(t) = \frac{2 \pi}{\hbar^2} \sum_{q^\prime} |V_{qq^\prime}|^2 \{ \langle N_q(t) \rangle + 1 \} \langle n^B_{q^\prime-q} \rangle - N_q(t) \langle n^B_{q-q^\prime} \rangle - \frac{2 \pi}{\hbar^2} \sum_{q^\prime} |V_{qq^\prime}|^2 \{ \langle N_q(t) \rangle + 1 \} \langle n^B_{q^\prime-q} \rangle - \frac{2 \pi}{\hbar^2} \sum_{q^\prime} |V_{qq^\prime}|^2 \{ \langle N_q(t) \rangle + 1 \} \langle n^B_{q^\prime-q} \rangle,
\end{equation}

where \( n^B_{q-q^\prime} \) and \( \Omega_q \) are the population and the frequency dispersion relation of the phonons in the thermal bath and \( V_{qq^\prime} \), the matrix element of the magnon-phonon interaction. After some mathematical handling, this Eq. (4) can be rewritten in the form

\begin{equation}
F_q(t) = \sum_{q^\prime} \chi_{qq^\prime} \{ \langle N_q(t) \rangle + 1 \} \langle n^B_{q^\prime-q} \rangle \cdot e^\beta q \cdot \langle n^B_{q^\prime-q} \rangle - N_q(t) \langle n^B_{q^\prime-q} \rangle \cdot e^\beta q \cdot \langle n^B_{q^\prime-q} \rangle,
\end{equation}

where

\begin{equation}
\chi_{qq^\prime} = \frac{2 \pi}{\hbar^2} |V_{qq^\prime}|^2 \{ \langle n^B_{q^\prime-q} \rangle \delta (\omega_q - \omega_{q^\prime} - \Omega_{q-q^\prime}) + e^\beta q \cdot \langle n^B_{q^\prime-q} \rangle \delta (\omega_q - \omega_{q^\prime} + \Omega_{q-q^\prime}) \cdot \nu_{q^\prime-q^\prime}^B.
\end{equation}

with this Eq. (5) having the form given originally by Fröhlich\textsuperscript{3,4} and we call it Fröhlich contribution. In the form of Eq. (4), it consists of two contributions: the first one on the right is positive and non-null for modes \( q^\prime \) in the sum for which \( \omega_q > \omega_{q^\prime} \). The other contribution is negative and non-null for modes \( q^\prime \) in the sum for which \( \omega_q < \omega_{q^\prime} \), representing a relaxation to the thermal bath. The balance of both contributions, as given by Eq. (5), is positive for \( \omega_q > \omega_{q^\prime} \) and negative for \( \omega_q < \omega_{q^\prime} \). Hence it plays a role of a pumping effect on mode \( q^\prime \) coming out of the modes \( q^\prime \) when \( \omega_q < \omega_{q^\prime} \). We stress that the expression contains nonlinear terms which are responsible for this behavior. Consequently, modes \( q^\prime \) for which \( \omega_q > \omega_{q^\prime} \) transfer their energy in excess of equilibrium to the mode \( q \) and therefore in a cascade down process it is transferred to the mode lowest in frequency. Thus, the mode lowest in frequency largely grows in population (drained from all the other modes) leading to the emergence of, what has been dubbed, a nonequilibrium Bose-Einstein condensation. Moreover, if we neglect the zero-point energy, \( F_q(t) \) of Eq. (4) becomes null, which means that it has a purely quantum-mechanical origin. Finally, we can summarize the point stating that such nonequilibrium Bose-Einstein condensation of “hot magnons” is of a pure quantum character and driven by Fröhlich nonlinear contribution to the kinetic equations, whose origin is in the interaction with the thermal bath in which the system is embedded (a description of the irreversible thermodynamics involved is presented in Ref. 6).

The other contributions in Eq. (3) are: (v) \( M_q(t) \) is the rate of change generated by the magnon-magnon interaction (exchange and dipolar), whose role is to lead the system of magnons to a state of internal thermalization. This contribution is in a “tug of war” with Fröhlich contribution (previous item); (vi) \( A_q(t) \) contains all the contributions coupling the populations to the amplitudes, \( \langle c_q(t) \rangle \) and \( \langle c_q^\dagger(t) \rangle \). Therefore this evolution equation is coupled to those of the other basic variables. The evolution equation for the amplitudes, also in a compact form, is in a linear approximation given by

\begin{equation}
\frac{d}{dt} \langle c_q(t) \rangle = - i \omega_q \langle c_q(t) \rangle - \Gamma_q(t) \langle c_q(t) \rangle,
\end{equation}

where, in Mori’s terminology,\textsuperscript{30} the first term on the right is a precession term and the second is in balance a relaxation (damping) term containing contributions arising out of the magnon-phonon interaction (of linear, Livshits and Fröhlich type in the nomenclature already introduced), of interaction with the radiation fields, and from the magnon-magnon interaction.
For numerical calculations and comparison with experiment, first we introduce a modeling consisting in a kind of “two-fluid model,” namely, we transform the large system of coupled evolution equations in a pair of coupled equations for the mean values of the variables over two regions of the Brillouin zone: one is a small region around the position of the minimum in energy in the dispersion relation, which we call $N_1$, and the other around the zone in which are the modes pumped by the rf fields, indicated by $N_2$. Besides that, in the conditions considered, pertaining to the experimental ones, it follows that the amplitudes $\epsilon_2(t)$ are small and rapidly decaying. Therefore, we proceed ahead with the evolution equation for the population decoupled from the amplitudes $[A_q(t)=0 \text{ in Eq. (3)}]$. For the mean populations $N_1(t)$ and $N_2(t)$ in the two-fluid model, we do have that

$$f_1 \frac{d}{dt} N_1(t) = -M(N_1(N_1+1) + N_2(N_2+1))\epsilon_1^2 N_1 - N_2 + F[N_1N_2 + (\bar{\nu}+1)N_2 - \bar{\nu}N_1] - f_1[N_1 - N_1^0] - D[N_1 - N_1^0]N_1,$$

and

$$f_2 \frac{d}{dt} N_2(t) = M(N_1(N_1+1) + N_2(N_2+1))\epsilon_2^2 N_2 - N_1 + F[N_1N_2 + (\bar{\nu}+1)N_2 - \bar{\nu}N_1] - f_2[N_2 - N_2^0] - D[N_2 - N_2^0]N_2 + (1 + 2N_2)I,$$

where $\bar{t}$ is the scaled time $t/\tau$, taking the relaxation time $\tau_q$ as having a unique constant value ($q$ independent), $N_1^0$ is the population in equilibrium, $\epsilon_1^0 = \epsilon_2^0 = N_2^0/N_1^0$, and $f_1$ and $f_2$ the fractions of the Brillouin zone corresponding to the two regions in the two-fluid model. Moreover, the coefficients $M$ and $F$ are the coupling strengths associated to magnon-magnon interaction and to Fröhlich contribution, respectively, $D$ is the one associated to decay with emission of photons and $\bar{\nu}$ is an average population of the phonons; the Livshits term can be neglected. Finally, $I$ stands for the intensity of the rf-radiation field transferred to the spin system, whose absorption is reinforced by a positive feedback effect. All these coefficients are addimensional, being multiplied by the relaxation time $\tau$.

Let us adopt the system of Eqs. (8) and (9) to understand the experiment in Ref. 1. The fraction $f_1$ follows considering the region of width 0.2 GHz (the indetermination in the experiment) around the minimum of frequency and $f_2$ is taken as corresponding to the region in the Brillouin zone around the pumping mode (4.1 GHz) with the same 0.2 GHz width. For YIG (Ref. 1) the transient time for attaining internal thermalization is about 200 ns and equating this with the linearized magnon-magnon term in Eq. (8) it gives $M \approx 10^{-13}$. $\tau$ is interpreted has the linear relaxation time to lattice, being on the order of a few microseconds (in numerical simulations we used $\tau=1 \mu s$). On the other hand, an estimation of the transient time for the steady state to set in allows to evaluate that $D \approx 10^{-11}$ and an analysis of the experimental data suggests that $F \approx 10^{-6}$.

Using the experimental data,1 varying the values of the parameters around those given above and adjusting the scaled intensity to $I=8 \times 10^{-4}$ (that, if $\tau=1 \mu s$, corresponds roughly to 0.5 W absorbed by the system), it follows the good agreement of theory and experiment shown in Fig. 1.

With the same parameters but considering constant application of the pumping source, the steady-state populations are obtained as a function of the source intensity, what is shown in Fig. 2. It can be noticed the existence of a critical
intensity (better saying, an intensity threshold) after which there follows a steep increase in population corresponding to the emergence of BEC. With increasing pumping intensity a second critical intensity (intensity threshold) is evidenced such that for higher values of $I$ is observed internal thermalization of the magnons which acquire a common quasi-temperature. This implies that the magnon-magnon interaction overcomes Fröhlich contribution and BEC is impaired.

In summary, as experimentally evidenced by Demokritov et al., the spin system in magnetic thin films under excitation by rf radiation, display a phenomenon of the type of a Bose–Einstein condensation of the hot magnons. The theoretical analysis performed here, in terms of a nonequilibrium statistical thermomechanics, shows that, in fact, this BEC of magnons follows in the way expected for many-boson systems embedded in a thermal bath as demonstrated by Fröhlich.$^{3,4}$

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