Statistical Thermodynamics of the Fröhlich-Bose-Einstein Condensation of Magnons out of Equilibrium

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(Dated: December 23, 2021)

A non-equilibrium statistical-thermodynamic approach to the study of a Fröhlich-Bose-Einstein condensation of magnons under radio-frequency radiation pumping is presented. Such a system displays a complex behavior consisting in steady-state conditions to the emergence of a synergetic dissipative structure resembling the Bose-Einstein condensation of systems in equilibrium. A kind of “two fluid model” arises: the “normal” non-equilibrium structure and Fröhlich condensate or “non-equilibrium” one, which is shown to be an attractor to the system. We analyze some aspects of the irreversible thermodynamics of this dissipative complex system, namely, its informational entropy, expressions for the fluctuations in non-equilibrium conditions, the associated Maxwell relations and the formulation of a generalized $\mathcal{H}$-theorem. We also study the informational entropy production of the system, an order parameter is introduced and Glansdorff-Prigogine criteria for evolution and (in)stability are verified.

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I. INTRODUCTION

It has been noticed [1] that the study of collective spin excitations in magnetically ordered materials (so-called spin waves and the associated quasi-particles, the magnons) has a successful history of more than 80 years [2], which recently has re-emerged within a young field of research and technology referred-to as Magnonics. This term Magnonics is considered to describe the sub-field of magnetic dynamic phenomena. The name Magnonics was created by analogy with Electronics, with the magnons acting in the transference of information instead of the electron charges in devices.

One important result pertaining to Magnonics has been the observation of a macroscopic quantum phenomenon resembling a Bose-Einstein condensation of magnons excited out of equilibrium by action of an electromagnetic field in the radio-frequency portion of the spectrum.

The kinetic of evolution of the system of spins in thin films of yttrium-iron-garnets (YIG) in the presence of a constant magnetic field, and being excited by a source of rf-radiation which drives the system towards far-removed from equilibrium conditions, has been reported in detailed experiments performed by Demokritov et al. [3, 4]. These experimental results have evidenced the occurrence of an unexpected large enhancement of the population of the magnons in the state lowest in energy in their energy dispersion relation. That is, the energy pumped on the system instead of being redistributed among the magnons in such non-thermal conditions is transferred to the mode lowest in frequency (with a fraction of course being dissipated to the surrounding media). Some theoretical studies along certain approaches has been presented by several authors (see for example Refs. [5–7]); we proceed here to describe the phenomenon within a complete thermo-statistical description within the framework of a non-equilibrium ensemble formalism.

Such phenomenon has been referred-to as a non-equilibrium Bose-Einstein condensation, which then would belong to a family of three types of BEC:

The original one is the BEC in many-boson particle systems in equilibrium at very low temperatures, which follows when their de Broglie thermal wave length becomes larger than their mean separation distance, and presenting some typical hallmarks (spontaneous symmetry breaking, long-range coherence, etc.). Aside from the case of superfluidity, BEC was realized in systems consisting of atomic alkali gases contained in traps. A nice tutorial review is due to A. J. Leggett [8] (see also [9]).

A second type of BEC is the one of boson-like quasi-particles, that is, those associated to elementary excitations in solids (e.g. phonons, excitons, hybrid excitations, etc.), when in equilibrium at extremely low temperatures. A well studied case is the one of an exciton-polariton system confined in microcavities (a near two-dimensional sheet), exhibiting the classic hallmarks of a BEC [10].

The third type, the one we are considering here, is the case of boson-like quasi-particles (associated to elementary excitations in solids) which are driven out of equilibrium by external perturbative sources. D. Snoke [11] has properly noticed that the name BEC can be misleading (some authors call it “resonance”, e.g. in the case of phonons [12]), and following this author it is better not to be haggling about names, and we introduce the nomenclature NEFBEC (short for Non-Equilibrium Fröhlich-Bose-Einstein Condensation for the reasons stated below). As noticed, here we consider the case of magnons (boson-like quasi-particles), demonstrating that NEFBEC of magnons 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 non-linear 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 medicine.

1. A first 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 [13-16]. From a Science, Technology
and Innovation (STI) point of view it was considered to have implications in medical diagnosis[17]. More recently has been considered to be related to brain functioning and artificial intelligence[18].

2. A second case is the one of acoustic vibration (ac phonons) in biological fluids, involving nonlinear anharmonic interactions and in the presence of pumping sonic waves, with eventual STI relevance in supersonic treatments and imaging in medicine[13, 20].

3. A third one is that of excitons (electron-hole pairs in semiconductors) interacting with the lattice vibrations and under the action of rf-electromagnetic fields; on a STI aspect, the phenomenon has been considered for allowing a possible exciton-laser in the THz frequency range called “Excitoner”[21, 22].

4. A fourth one is the case of magnons already referred to[3, 4], which we here analyze in depth. The thermal bath is constituted by the phonon system, with which a nonlinear interaction exists, and the magnons are driven arbitrarily out of equilibrium by a source of electromagnetic radio frequency[23, 24]. Technological applications are related to the construction of sources of coherent microwave radiation[25, 26].

There exist two other cases of NEBEC (differing from NEFBEC) where the phenomenon is associated to the action of the pumping procedure of drifting electron excitation, namely,

5. A fifth one consists in a system of longitudinal acoustic phonons driven away from equilibrium by means of drifting electron excitation (presence of an electric field producing an electron current), which has been related to the creation of the so-called Saser, an acoustic laser device, with applications in computing and imaging[27, 28].

6. A sixth one involving a system of LO-phonons driven away from equilibrium by means of drifting electron excitation, which displays a condensation in an off-center small region of the Brillouin Zone[29, 30].

We describe here item number 4, namely, a system of magnons excited by an external pumping source. For that purpose, we 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) which is considered to be in equilibrium with an external reservoir at temperature \( T_0 \). The microscopic state of the system is characterized by the full Hamiltonian of spins and lattice vibrations after going through Holstein-Primakov and Bogoliubov transformations[31–33]. On the other hand, the characterization of the macroscopic state of the magnon system is done in terms of the Thermo-Mechanical Statistics based on the framework of a Non-Equilibrium Statistical Ensemble Formalism (NESEF for short)[34–39]. Other modern approach consists in the use of Computational Modeling[40, 41] (developed after Non-equilibrium Molecular Dynamics[42]). It may be noticed that NESEF is a systematization and an extension of the essential contributions of several renowned authors following the brilliant pioneering work of Ludwig Boltzmann. The formalism introduces the fundamental properties of historicity and irreversibility in the evolution of the non-equilibrium system where dissipative and pumping processes are under way.

In terms of the dynamics generated by the full Hamiltonian the equations of evolution of the macroscopic state of the system are obtained in the framework of the NESEF-based nonlinear quantum kinetic theory[34–39, 43–46]. We call the attention to the fact that the evolution equations are the quantum mechanical equations of motion averaged over the non-equilibrium ensemble, with the NESEF-kinetic theory providing a practical way of calculation. The evolution of the non-equilibrium state of magnons under rf-radiation excitation is fully described in Refs. [23, 24] (for the sake of completeness we summarize the results in Section II), and, on the basis of it we present here a extended study of the non-equilibrium irreversible thermodynamics of
the Fröhlich-Bose-Einstein condensation of such “hot” magnons. This is done in terms of the NESEF-based Nonequilibrium-Statistical Irreversible Thermodynamics [47, 48] (also Ch. 7 in Ref. [34]).

II. FRÖHLICH-BOSE-EINSTEIN CONDENSATION OF HOT MAGNONS IN BRIEF [23, 24]

The system we are considering consists of a subsystem of spin $s$ being pumped by a microwave source and interacting non-linearly with a thermal bath (black-body radiation and crystalline lattice) that is in contact with a thermal reservoir in equilibrium at temperature $T_0$. This system is well described by the Hamiltonian

$$\hat{H} = \hat{H}_S + \hat{H}_Z + \hat{H}_{SR} + \hat{H}_{SL} + \hat{H}_L,$$

where $\hat{H}_S$ accounts for the internal (exchange and magnetic dipole) interactions between spins, $\hat{H}_Z$ is associated with the effect of the constant magnetic field (Zeeman Effect). $\hat{H}_L$ and $\hat{H}_R$ are the Hamiltonian of the thermal bath (lattice and radiation respectively), $\hat{H}_{SL}$ and $\hat{H}_{SR}$ their interaction with the spin subsystem ($\hat{H}_{SR}$ includes also the effect of the source). Introducing the quasi-particles related to the spin, lattice and radiation variables (respectively the magnons, phonons and photons) and their creation and annihilation operators ($\hat{c}^\dagger_q, \hat{c}_q, \hat{b}_k^\dagger, \hat{b}_k, d_q^\dagger$ and $d_q$) we may write the Hamiltonian of Eq. (1) as

$$\hat{H} = \hat{H}_0 + \hat{H}',$$

with

$$\hat{H}_0 = \hat{H}_S^{(2)} + \hat{H}_L + \hat{H}_R =$$

$$= \sum_q \hbar \omega_q \hat{c}^\dagger_q \hat{c}_q + \sum_k \hbar \Omega_k \hat{b}_k^\dagger \hat{b}_k + \sum_p \hbar \zeta_p \hat{d}_p^\dagger \hat{d}_p$$

being the non-interacting term formed by the Hamiltonians of free magnons, phonons and photons, and $\hbar \omega_q, \hbar \Omega_k$ and $\hbar \zeta_p$ their energies. The other term,

$$\hat{H}' = \hat{H}_{MM} + \hat{H}_{SL} + \hat{H}_{SR},$$

includes the interactions between quasi-particles:

$$\hat{H}_{MM} = \sum_{q_1, q_2} \mathcal{V}_{q_1, q_2} \hat{c}^\dagger_{q_1} \hat{c}_{q_1} \hat{c}_{q_2} \hat{c}_{q_2+q_1-q_2}$$

is the magnon-magnon scattering term;

$$\hat{H}_{SL} = \sum_{q, k \neq 0} (\hat{b}_k^\dagger + \hat{b}_k) \left\{ \mathcal{F}_{q, k} \hat{c}_{q-k}^\dagger \hat{c}_{q-k} + \mathcal{L}_{q, k} \hat{c}_{q-k}^\dagger \hat{c}_{q-k}^\dagger + \mathcal{L}_{q, -k} \hat{c}_q \hat{c}_{q-k}^\dagger \hat{c}_{q-k} + \mathcal{L}_{q, k} \hat{c}_q \hat{c}_{q-k} \hat{c}_{q-k}^\dagger \right\} +$$

$$+ \sum_{q, k \neq 0} \left\{ \mathcal{R}_{q, k} \hat{b}_q^\dagger \hat{b}_k - \hat{b}_q^\dagger \hat{b}_q - \hat{b}_k^\dagger \hat{b}_k \right\} (\hat{c}_q + \hat{c}_{-q}^\dagger)$$
accounts for the relevant magnon-phonon interaction and
\[ \mathcal{H}_{SR} = \sum_p (\hat{d}_p + \hat{d}^\dagger_p) \left( \mathcal{S}^l_p \hat{c}_p \mathcal{S}^r_p \hat{c}^\dagger_p + \mathcal{S}^l_p \hat{c}^\dagger_p \mathcal{S}^r_p \hat{c}_p \right) + \]
\[ + \sum_{p,q} (\hat{d}_p + \hat{d}^\dagger_p) \left( \mathcal{S}^{lb}_{q,p} \hat{c}_{q-p} \mathcal{S}^{rb}_{q,p} \hat{c}^\dagger_{q-p} + \mathcal{S}^{lb}_{q,p} \hat{c}^\dagger_{q-p} \mathcal{S}^{rb}_{q,p} \hat{c}_{q-p} \right) \]  
(6)
is the interaction between magnons and photons (source and black-body radiation).

After the mechanical description of the system follows the thermodynamical one. The thermodynamical state can be defined in terms of the time-dependent thermodynamical variables
\[ \left\{ \left\{ \mathcal{N}_q(t) \right\}; \left\{ \mathcal{N}_{q,Q}(t) \right\}; \left\{ \langle \hat{c}^\dagger_q t \rangle \right\}; \left\{ \langle \hat{c}_q t \rangle \right\}; \left\{ \sigma^q(t) \right\}; \left\{ \sigma_{q,Q}(t) \right\}; \left\{ E_B \right\}, \right. \]
(7)

average values of the so-called basic micro-dynamical variables
\[ \left\{ \left\{ \hat{N}_q \right\}; \left\{ \hat{N}_{q,Q} \right\}; \left\{ \hat{c}^\dagger_q \right\}; \left\{ \hat{c}_q \right\}; \left\{ \hat{\sigma}^q \right\}; \left\{ \hat{\sigma}_{q,Q} \right\}; \left\{ \hat{\sigma}_{q,Q} \right\}; \left\{ \hat{\sigma}_{q,Q} \right\}; \left\{ \hat{\tau}_{q,Q} \right\} \right. \],
(8)

with \( Q \neq 0 \), where
\[ \hat{\mathcal{H}}_B = \hat{\mathcal{H}}_L + \hat{\mathcal{H}}_R, \]
(9)
is the Hamiltonian of the thermal bath (phonons and photons),
\[ \hat{N}_q = \hat{c}^\dagger_q \hat{c}_q, \quad \hat{N}_{q,Q} = \hat{c}^\dagger_{q+\mathcal{Q}} \hat{c}_{q-\mathcal{Q}}, \]
(10)
with \( \hat{N}_q \) being the population operator of magnons in mode \( q \), \( \hat{N}_{q,Q} \) describing its change in space (inhomogeneities in populations) and we recall that \( \hat{c}^\dagger_q \) (\( \hat{c}_q \)) are single-magnons operators whose eigenstates are the coherent states. Finally,
\[ \hat{\sigma}^q = \hat{c}^\dagger_q \hat{c}_q, \quad \hat{\sigma}_{q,Q} = \hat{c}^\dagger_{q-\mathcal{Q}} \hat{c}^\dagger_{q-\mathcal{Q}} \]
(11)
are the Hugenholtz-Gorkov pairs of two magnons.

These averages are weighted through a non-equilibrium statistical operator \( \hat{\mathcal{N}}_c(t) \), for example, \( \mathcal{N}_q(t) = \text{Tr} \left\{ \hat{N}_q \hat{\mathcal{N}}_c(t) \right\} \). We introduce a factorization between the thermal bath in equilibrium and the magnetic subsystem
\[ \hat{\mathcal{N}}_c(t) = \hat{\rho}_c(t) \times \hat{\rho}_B, \]
(12)
where
\[ \hat{\rho}_B = \frac{1}{Z_B} \exp \left\{ -\beta_B \left( \mathcal{H}_B \right) \right\} \]
(13)
is the canonical distribution function of the phonons and photons in stationary condition near equilibrium at temperature \( T_B = (k_B \beta_B)^{-1} \) (being \( Z_B \) its partition function) and \( \hat{\rho}_c(t) \) is the
It is important here to make three observations: first, we stress that the auxiliary statistical operator has the form of an instantaneous generalized canonical distribution that tends to the canonical one when the system is in equilibrium with all intensive non-equilibrium thermodynamic variables for example $N_q(t) = \text{Tr} \left\{ \hat{N}_q \hat{\rho}_q(t) \right\} = \text{Tr} \left\{ \hat{N}_q \hat{\rho}_q(t) \times \hat{\rho}_B \right\}$. Second, the expression adopted in Eq. (15) for the statistical operator incorporates the dynamical evolution while, on the other hand, includes irreversibility [34–36].

The auxiliary statistical operator $\hat{\rho}_q(t)$ is written in terms of the chosen micro-dynamical variables taken the form

$$\hat{\rho}_q(t) = \frac{1}{Z(t)} \exp \left\{ -\sum_q \left[ F_q(t) \hat{N}_q + \phi_q(t) \hat{c}_q + \phi^*_q(t) \hat{c}_q^\dagger + \varphi_q(t) \hat{\sigma}_q + \varphi^*_q(t) \hat{\sigma}_q^\dagger \right] - \sum_{q,Q} \left[ F_{q,Q}(t) \hat{N}_{q,Q} + \varphi_{q,Q}(t) \hat{\sigma}_{q,Q} + \varphi^*_{q,Q}(t) \hat{\sigma}^\dagger_{q,Q} \right] \right\},$$

where

$$\left\{ \{ F_q(t) \}; \{ F_{q,Q}(t) \}; \{ \phi_q(t) \}; \{ \phi^*_q(t) \}; \{ \varphi_q(t) \}; \{ \varphi^*_q(t) \}; \{ \varphi_{q,Q}(t) \}; \{ \varphi^*_{q,Q}(t) \} \right\},$$

are the non-equilibrium thermodynamic variables conjugated to the basic variables contained in set (7) in the sense of the Eqs. (18) and (19) below. The normalization of $\hat{\rho}(t,0)$ introduces the non-equilibrium partition function

$$\tilde{Z}(t) \equiv \text{Tr} \left\{ \exp \left\{ -\sum_q \left[ F_q(t) \hat{N}_q + \phi_q(t) \hat{c}_q + \phi^*_q(t) \hat{c}_q^\dagger + \varphi_q(t) \hat{\sigma}_q + \varphi^*_q(t) \hat{\sigma}_q^\dagger \right] - \sum_{q,Q} \left[ F_{q,Q}(t) \hat{N}_{q,Q} + \varphi_{q,Q}(t) \hat{\sigma}_{q,Q} + \varphi^*_{q,Q}(t) \hat{\sigma}^\dagger_{q,Q} \right] \right\} \right\}.$$  

It is important here to make three observations: first, we stress that the auxiliary statistical operator $\hat{\rho}(t,0)$ does not describe the irreversible time-evolution of the system, and the average values weighted with $\hat{\rho}(t,0)$ coincide with those weighted with $\hat{\rho}_q(t)$ only for the micro-dynamical variables, for example $N_q(t) = \text{Tr} \left\{ \hat{N}_q \hat{\rho}_q(t) \right\} = \text{Tr} \left\{ \hat{N}_q \hat{\rho}_q(t) \times \hat{\rho}_B \right\}$. Second, the expression adopted in Eq. (15) for the statistical operator has the form of an instantaneous generalized canonical distribution that tends to the canonical one when the system is in equilibrium with all the present intensive variables except $F_q(t)$ (that, in this case, is associated with the magnons equilibrium temperature) going to zero. Finally, since the intensive non-equilibrium thermodynamic variables of set (16) equivalently describe the macro-state of the system and that

$$\frac{\delta \ln \tilde{Z}(t)}{\delta \phi_q(t)} = \langle \hat{c}_q^\dagger \rangle, \quad \frac{\delta \ln \tilde{Z}(t)}{\delta F_q(t)} = N_q(t), \quad \frac{\delta \ln \tilde{Z}(t)}{\delta \varphi_q(t)} = \sigma_q(t),$$

non-equilibrium statistical operator of the magnon system. The last one may be obtained solving a modified Liouville-Dirac equation for $\hat{\rho}_q(t)$,

$$-\frac{\partial}{\partial t} \hat{\rho}_q(t) + \frac{1}{i\hbar} \left[ \hat{\rho}_q(t), \hat{H} \right] = -\varepsilon \left\{ \hat{\rho}_q(t) - \hat{\rho}(t,0) \right\},$$

where the right term (with $\varepsilon \to 0$) introduces the “Bogoliubov’s symmetry-breaking procedure” in time and $\hat{\rho}(t,0)$ is the auxiliary statistical operator. Equation (14) ensures on the one hand that the non-equilibrium statistical operator $\hat{\rho}_q(t)$ incorporates the dynamical evolution while, on the other hand, includes irreversibility [34–36].
\[
- \frac{\delta \ln \bar{Z}(t)}{\delta F_{q,Q}(t)} = N_{q,Q}(t), \quad - \frac{\delta \ln \bar{Z}(t)}{\delta \varphi_{q,Q}(t)} = \sigma_{q,Q}(t),
\] (19)

may be considered non-equilibrium equations of state, there is a close analogy with the intensive thermodynamic variables in equilibrium.

After presenting the relevant variables and the non-equilibrium statistical operator, the next step in the thermodynamical description is the derivation of the evolution equations of the thermodynamical variables in set (7). Such equations form a system of nonlinear coupled integro-differential equations which is discussed in Ref. [23] and in a detailed form in Ref. [24]. As stated there, for specific spin systems, it suffices to follow the evolution of magnons’ populations \( \{ N_{q}(t) \} \); moreover for the equation of state it follows that
\[
\langle \hat{c}_{q}^{\dagger} \hat{c}_{q} \rangle_{t} = N_{q}(t) = \frac{1}{e^{F_{q}(t)} - 1},
\] (20)
or, alternatively,
\[
F_{q}(t) = \ln \left\{ 1 + \frac{1}{N_{q}(t)} \right\} = - \ln \left\{ \frac{N_{q}(t)}{N_{q}(t) + 1} \right\},
\] (21)

We recall that the equations of evolution for the populations are the quantum mechanical equations of motion for the dynamical quantities \( \hat{N}_{q} \) averaged over the non-equilibrium ensemble. They are handled resorting to the NESEF-based nonlinear quantum kinetic theory, with the calculations performed in the approximation that incorporates only terms quadratic in the interaction strength - with memory and vertex renormalization neglected, that is, we keep what in kinetic theory is called the irreducible part of the two-particle collisions -
\[
\frac{d}{dt} N_{q}(t) = \frac{1}{i\hbar} \text{Tr} \left\{ \left[ \hat{N}_{q}, \hat{H} \right] \hat{\rho}(t) \times \hat{\rho}_{B} \right\} = J_{N_{q}}^{(0)}(t) + J_{N_{q}}^{(1)}(t) + J_{N_{q}}^{(2)}(t),
\] (22)
\[
J_{N_{q}}^{(0)}(t) = \frac{1}{i\hbar} \text{Tr} \left\{ \left[ \hat{N}_{q}, \hat{H}_{0} \right] \hat{\rho}(t) \times \hat{\rho}_{B} \right\} = 0,
\] (23)
\[
J_{N_{q}}^{(1)}(t) = \frac{1}{i\hbar} \text{Tr} \left\{ \left[ \hat{N}_{q}, \hat{H}' \right] \hat{\rho}(t) \times \hat{\rho}_{B} \right\} = 0,
\] (24)
\[
J_{N_{q}}^{(2)}(t) \approx J_{N_{q}}^{(2)}(t) = \frac{1}{(i\hbar)^{2}} \int_{-\infty}^{t} dt' e^{i(t'-t)} \text{Tr} \left\{ \left[ \hat{H}'(t'-t), [\hat{H}', \hat{N}_{q}] \right] \hat{\rho}(t) \times \hat{\rho}_{B} \right\} + \frac{1}{i\hbar} \sum_{\ell} \int_{-\infty}^{t} dt' e^{i(t'-t)} \text{Tr} \left\{ \left[ \hat{H}'(t'-t), \hat{P}_{\ell} \right] \hat{\rho}(t) \times \hat{\rho}_{B} \right\} \frac{\delta J_{N_{q}}^{(1)}(t)}{\delta Q_{\ell}(t)},
\] (25)
with \( \hat{P}_{\ell} \) and \( \hat{Q}_{\ell} \) being the variables of sets (3) and (7) respectively, and
\[
\hat{O}(t) = e^{-\frac{\hat{H}_{0}}{i\hbar}} \hat{O} e^{\frac{\hat{H}_{0}}{i\hbar}},
\] (26)
\( \delta \) stands for functional differentiation.
In a compact form we may write
\[
\frac{d}{dt} N_q(t) = \mathcal{S}_q(t) + \mathcal{R}_q(t) + L_q(t) + \mathfrak{S}_q(t) + \mathcal{M}_q(t),
\]  
(27)
where
\[
\mathcal{S}_q(t) = \frac{8\pi}{\hbar^2} \sum_{q' \neq -q} |S_{q,q+q'}^{b}|^2 \left\{ \left( 1 + N_{q+q'} \right) f_{q+q'}^S \right\} \delta(\omega_q + \omega_{q'} - \Omega_{q+q'})
\]
(28)
is the source term that accounts for the pumping of energy to the system, \( f_{q+q'}^S \) stands for the population of photons of the source;
\[
\mathcal{R}_q(t) = \frac{8\pi}{\hbar^2} \sum_{q' \neq -q} |S_{q,q+q'}^{b}|^2 \left\{ \left( N_{q+q'} + 1 \right) \left( N_q + 1 \right) f_{q+q'}^T - N_{q+q'} N_q f_{q+q'}^T \right\} \delta(\omega_q + \omega_{q'} - \zeta_{q+q'})
\]
(29)
is a nonlinear term of interaction between the spin subsystem and the black-body radiation (\( f_{q+q'}^T \) being its photon’s population);
\[
L_q(t) = -\frac{1}{\tau_q} \left[ N_q - N_q^{(0)} \right]
\]
(30)
is the linear relaxation to the lattice with characteristic time \( \tau_q \). The last two terms are nonlinear contributions;
\[
\mathfrak{S}_q(t) = \frac{2\pi}{\hbar^2} \sum_{q' \neq -q} |F_{q,q+q'}|^2 \left\{ \left( N_{q+q'} + 1 \right) \left( \nu_{q'} - q' + 1 \right) - \left( N_q + 1 \right) \nu_q - q \right\} \delta(\omega_q - \omega_{q'} - \Omega_{q+q'}) +
\]
\[
+ \frac{2\pi}{\hbar^2} \sum_{q' \neq -q} |F_{q,q+q'}|^2 \left\{ \left( N_{q+q'} + 1 \right) N_q \nu_q - q - N_q \left( \nu_{q+q'} + 1 \right) \left( \nu_{q+q'} - 1 \right) \right\} \delta(\omega_q - \omega_{q'} + \Omega_{q+q'})
\]
(31)
the so-called Fröhlich term, a nonlinear interaction between magnons mediated by the lattice, and
\[
\mathcal{M}_q(t) = \frac{16\pi}{\hbar^2} \sum_{q_1,q_2,q_3 \neq -q} |V_{q_1,q_2,q_3}|^2 \left\{ \left( N_{q_1} + 1 \right) N_{q_2} N_{q_3} - N_{q_1} \left( N_{q_2} + 1 \right) N_{q_3} \right\} \times \delta(\omega_{q_1} + \omega_{q_2} - \omega_{q_3}) \delta_{q_1,q_2+q_3,q_1-q_2}
\]
(32)
accounts for the magnon-magnon scattering interaction term.

Although the kinetic equations for the populations [Eq. (27)] may well describe the thermodynamic evolution of the magnetic subsystem, the complete thermodynamic description of the entire system must also include the evolution of the energy of the thermal bath \( E_B(t) \) (lattice and black-body radiation). In a similar form of Eq. (22) we have that
\[
\frac{d}{dt} E_B(t) = \frac{1}{i\hbar} \text{Tr} \left[ \left[ \hat{H}_B, \hat{\rho}_B(t) \right] \right] \simeq J_{E_B}^{(0)}(t) + J_{E_B}^{(1)}(t) + J_{E_B}^{(2)}(t).
\]
(33)
It is simple to show that \( J_{E_B}^{(0)}(t) \) and \( J_{E_B}^{(1)}(t) \) are null. The last term is composed by two contributions:
\[
J_{E_B}^{(2)}(t) = J_{T_B}^{(2)}(t) - \sum_q \hbar \omega_q \left[ \mathcal{M}_q(t) + L_q(t) + \mathfrak{S}_q(t) \right],
\]
(34)
the first,

\[ J_{TD}^{(2)}(t) = -\frac{E_B(t) - E_B^{(0)}}{\tau_{TD}}, \]  

is the contribution which accounts for the thermal diffusion to the reservoir with a thermal diffusion time \( \tau_{TD} \) and tends to lead the thermal bath to equilibrium (characterized by the equilibrium energy \( E_B^{(0)} \)). The other contribution is related to the energy received from the subsystem of magnons.

Our system has its thermodynamical evolution described by the kinetic equations (27) and (33) and they must be solved. Since we stated before that the thermal bath is in a stationary state near the equilibrium condition defined by the external reservoir we have that

\[ \frac{d}{dt} E_B(t) = J_{EU}^{(2)}(t) - \sum_q \hbar \omega_q [\mathcal{R}_q(t) + L_q(t) + \mathcal{F}_q(t)] = 0, \]

and the thermal diffusion effect is sufficiently rapid for keeping this configuration. In this case \( E_B(t) \simeq E_B^{(0)}, \ T_B \simeq T_0 \) and \( \beta_B \simeq \beta_0 \).

Considering again the evolution of the population of magnons, we emphasise that Eq. (27) constitutes a nonlinear system of coupled integro-differential equations. Its resolution in an approximate form called “two fluid model” is discussed on Refs. [23, 24], where the mean populations \( \mathcal{N}_1(t) \) and \( \mathcal{N}_2(t) \) were defined representing the populations of magnons around the minimum of frequency and those being fed by the external source respectively,

\[ \mathcal{N}_{1,2}(t) = \frac{\sum_{q \in R_{1,2}} N_q(t)}{\sum_{q \in R_{1,2}} N_q(t)_{1,2}}, \]  

\( R_1 \) and \( R_2 \) are the correspondent regions in the reciprocal space. Their evolution equations were obtained from Eq. (27),

\[ f_1 \frac{d}{dt} \mathcal{N}_1(\tilde{t}) = -D \mathcal{N}_1(\mathcal{N}_1 - \mathcal{N}_1^{(0)}) - f_1 \left[ \mathcal{N}_1 - \mathcal{N}_1^{(0)} \right] + \]  

\[ + F \{ \mathcal{N}_1 \mathcal{N}_2 + (\bar{\nu} + 1) \mathcal{N}_2 - \bar{\nu} \mathcal{N}_1 \} - \]  

\[ - M \{ \mathcal{N}_1 (\mathcal{N}_1 + 1) + \mathcal{N}_2 (\mathcal{N}_2 + 1) \} (\frac{\mathcal{N}_1^{(0)}}{\mathcal{N}_1} - \mathcal{N}_2), \]  

and

\[ f_2 \frac{d}{dt} \mathcal{N}_2(\tilde{t}) = 1(1 + 2 \mathcal{N}_2) - \]  

\[ - D \mathcal{N}_2(\mathcal{N}_2 - \mathcal{N}_2^{(0)}) - f_2 \left[ \mathcal{N}_2 - \mathcal{N}_2^{(0)} \right] - \]  

\[ - F \{ \mathcal{N}_1 \mathcal{N}_2 + (\bar{\nu} + 1) \mathcal{N}_2 - \bar{\nu} \mathcal{N}_1 \} + \]  

\[ + M \{ \mathcal{N}_1 (\mathcal{N}_1 + 1) + \mathcal{N}_2 (\mathcal{N}_2 + 1) \} (\frac{\mathcal{N}_2^{(0)}}{\mathcal{N}_2} - \mathcal{N}_2). \]  

where \( \tilde{t} \) is the scaled time \( t/\tau \), taking the relaxation time \( \tau_q \) as having a unique constant value
\( q \)-independent), \( \mathcal{N}_{1,2}^{(0)} \) are the populations in equilibrium, 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. Finally, the parameter \( I \) is related to the rate of the rf-radiation field transferred to the spin system, whose absorption is reinforced by a positive feedback effect. All these coefficients are dimensionless, being multiplied by the relaxation time \( \tau \).

In a similar fashion, the energy of the thermal bath has an evolution given, in the two fluid model, by

\[
\frac{d}{dt}E_B(t) = \tau J_{TD}(t) + \frac{\hbar}{2} \omega_1 \left( D_N_1 (N_1 - N_1^{(0)}) + f_1 \left( N_1 - N_1^{(0)} \right) - F \left( N_1 N_2 + (\bar{\nu} + 1) N_2 - \bar{\nu} N_1 \right) \right) + \frac{\hbar}{2} \omega_2 \left( D_N_2 (N_2 - N_2^{(0)}) + f_2 \left( N_2 - N_2^{(0)} \right) + F \left( N_1 N_2 + (\bar{\nu} + 1) N_2 - \bar{\nu} N_1 \right) \right),
\]

being \( \hbar \omega_1 \) and \( \hbar \omega_2 \) the energy of the magnons in the regions \( R_1 \) and \( R_2 \), and \( n = \sum_{q} 1 \).

On Fig. 1 we show the evolution of the populations \( N_1 \) and \( N_2 \), departing from equilibrium, under the action of the pumping source (we adopted \( \tau = 1 \mu s \) for comparison with experimental data \([3]\)), solving numerically Eqs. (38) and (39). As stated on Refs. \([23, 24]\), besides the good agreement with the experimental data, this result shows clearly the accumulation of magnons on the mode of minimum frequency (\( N_1 \)).

Moreover, the analysis of the steady state of the system, i. e., the solutions of Eqs. (38) and (39) such that \( \frac{d}{dt}N_1(t) \) and \( \frac{d}{dt}N_2(t) \) are null, make evident the role of the Fröhlich term to the condensation of magnons. On Fig. 2 we show the values of the steady-state populations, \( \mathcal{N}_1^S \) and \( \mathcal{N}_2^S \) as a function of the scaled rate of pumping \( I \), and it can be noticed the existence of two pumping scaled rate thresholds, the first, after which there follows a steep increase in the population of the modes lowest in frequency, corresponds to the emergence of BEC, while the
second, for higher values of \( I \), accounts for the internal thermalization of the magnons which acquire a common quasi-temperature, implying that the magnon-magnon interaction overcomes Fröhlich contribution and BEC is impaired.

The stability of these solutions was analyzed firstly through the evaluation of the Lyapunov exponents. Defining the variables \( a, b, c \) and \( d \) in such manner that

\[
\frac{\partial}{\partial N_1} \frac{d}{dt} f_1 N_1(t) = -D(2N_1^* - N_1^{(0)}) - f_1 + F \{ N_2^* - \bar{\nu} \} + M (2N_1^* + 1) N_2^* - \\
- M \frac{N_2^{(0)}}{N_1^{(0)}} \{ N_1^* (3N_1^* + 2) + N_2^* (N_2^* + 1) \} \equiv a f_1 
\]

\[
\frac{\partial}{\partial N_2} \frac{d}{dt} f_1 N_1(t) = F \{ N_1^* + \bar{\nu} + 1 \} + M \{ N_1^* (N_1^* + 1) + N_2^* (3N_2^* + 2) \} - \\
- M \frac{N_2^{(0)}}{N_1^{(0)}} \{ N_1^* (2N_2^* + 1) \} \equiv b f_1, 
\]

\[
\frac{\partial}{\partial N_1} \frac{d}{dt} f_2 N_2(t) = - F \{ N_2^* - \bar{\nu} \} + M \frac{N_2^{(0)}}{N_1^{(0)}} \{ N_1^* (3N_1^* + 2) + N_2^* (N_2^* + 1) \} - \\
- M (2N_1^* + 1) N_2^* \equiv c f_2, 
\]

\[
\frac{\partial}{\partial N_2} \frac{d}{dt} f_2 N_2(t) = 2I - D(2N_2^* - N_2^{(0)}) - f_2 - F \{ N_1^* + \bar{\nu} + 1 \} + \\
+ M \frac{N_2^{(0)}}{N_1^{(0)}} \{ N_1^* (2N_2^* + 1) \} - \\
- M \{ N_1^* (N_1^* + 1) + N_2^* (3N_2^* + 2) \} \equiv d f_2, 
\]

we have that the Lyapunov exponents are the solutions of

\[
\begin{vmatrix}
  a - \lambda & b \\
  c & d - \lambda
\end{vmatrix} = (a - \lambda)(d - \lambda) - bc = 0, 
\]

or
where, we recall, \( \hat{\mathcal{R}}_\varepsilon(t) \) is the non-equilibrium statistical operator of Eq. (12) and \( \mathcal{P}_\varepsilon(t) \) is a time-dependent projection operator (it is characterized by the non-equilibrium state of the system at any time \( t \)) such that \[ \mathcal{P}_\varepsilon(t) \ln \hat{\rho}_\varepsilon(t) = \ln \hat{\rho}(t,0), \]
and
\[ \mathcal{P}_N(t) \ln \dot{\rho}_B = \ln \dot{\rho}_B, \quad (50) \]
where \( \dot{\rho}(t, 0) \) and \( \dot{\rho}_B \) are those of Eqs. (15) and (13) but in the contracted description that takes as relevant micro-variables, as stated before, only the occupation-number operator of magnons, \( \hat{N}_q \) and the Hamiltonian of the thermal bath, \( \hat{H}_B \), that is, in Eq. (15) the terms involving \( \phi_q, \phi_q, F_q \), and their conjugates are neglected.

Hence we have that
\[ S(t) = -\text{Tr} \left\{ \hat{R} \left( \ln \{ \hat{\rho}(t, 0) \times \hat{\rho}_B \} \right) \right\} = \phi(t) + \beta_B E_B + \sum_q F_q(t) \hat{N}_q(t), \quad (51) \]
\[ \phi(t) = \ln Z_B + \ln \bar{Z}(t), \quad (52) \]
where \( Z_B \) and \( \bar{Z}(t) \) are the canonical and non-equilibrium partition functions [see Eqs. (13) and (17)]. The last one depends on time and must be explicitly written in terms of the non-equilibrium thermodynamic variables. In a analogous way to the equilibrium Bose-statistics we obtain that
\[ \bar{Z}(t) = \text{Tr} \exp \{- \sum_q F_q(t) \hat{N}_q \}, \]
\[ \ln \bar{Z}(t) = \sum_q \ln \frac{1}{1 - e^{-F_q(t)}}. \]

Thus
\[ \ln \bar{Z}(t) = \sum_q \ln \frac{1}{1 - e^{-F_q(t)}} \]
and using the relation between \( F_q(t) \) and \( \hat{N}_q(t) \), Eq. (21), one obtains the expression for the informational entropy
\[ S(t) = \ln Z_B + \beta_B E_B - \sum_q \{ \hat{N}_q(t) \ln [\hat{N}_q(t)] - [\hat{N}_q(t) + 1] \ln [\hat{N}_q(t) + 1] \}, \]
whereas \( Z_B, \beta_B \) and \( E_B \) are constants [see Eq. (36) and subsequent discussion].

B. Fluctuations and Maxwell Relations

As already shown, the average value of any dynamical quantity of the basic set in NESEF is given by minus the functional derivative of the generating functional \( \phi(t) \) with respect to the associated non-equilibrium thermodynamic variables [and we recall that this function can be related to a kind of non-equilibrium partition function through the expression \( \phi(t) = \ln Z_B + \ln \bar{Z}(t) \), cf. Eqs. (17), (18), (19) and (52)]. Considering only the populations of magnons as relevant variables,
\[ \bar{Z}(t) = \text{Tr} \exp \left\{ -\sum_q F_q(t) \hat{N}_q \right\}, \]
\[ \bar{S}(t) = \ln Z_B + \beta_B E_B - \sum_q \{ \hat{N}_q(t) \ln [\hat{N}_q(t)] - [\hat{N}_q(t) + 1] \ln [\hat{N}_q(t) + 1] \}, \]
and the matrix is symmetrical, that is, square deviations, or fluctuations, of quantities matrix of correlations

\[ C \]

where components of minus the inverse of the matrix of correlations \( C \)

Moreover, we find that

\[ \Delta N_q' = \hat{N}_q' - \text{Tr} \left\{ \hat{N}_q' \hat{\theta}(t, 0) \right\} = \hat{N}_q' - N_q(t), \tag{59} \]

and Eq. (58) defines the matrix of correlations \( \hat{C}(t) \). Their diagonal elements are the mean square deviations, or fluctuations, of quantities \( \hat{N}_q' \), namely

\[ C_{qq}(t) = \text{Tr} \left\{ \left[ \Delta \hat{N}_q \right]^2 \hat{\theta}(t, 0) \right\} = \text{Tr} \left\{ \left[ \hat{N}_q - N_q(t) \right]^2 \hat{\theta}(t, 0) \right\} = \Delta^2 N_q(t) \tag{60} \]

and the matrix is symmetrical, that is,

\[ C_{q'q''}(t) = \frac{\delta^2 \phi(t)}{\delta F_{q'}(t) \delta F_{q''}(t)} = \frac{\delta^2 \phi(t)}{\delta F_{q'}(t) \delta F_{q''}(t)} = C_{q''q'}(t) \tag{61} \]

what is a manifestation in IST of the known Maxwell relations in equilibrium.

Let us next scale the informational entropy and the non-equilibrium thermodynamic intensive variables in terms of Boltzmann constant, \( k_B \), that is, we introduce

\[ S(t) = k_B \hat{S}(t); \quad F_q(t) = k_B F_q(t); \tag{62} \]

and then, because of Eq. (51),

\[ F_q(t) = \frac{\delta \hat{S}(t)}{\delta \hat{N}_q(t)}. \tag{63} \]

Moreover, we find that

\[ \frac{\delta^2 \hat{S}(t)}{\delta \hat{N}_q'(t) \delta \hat{N}_{q''}(t)} = \frac{\delta F_{q'}(t)}{\delta \hat{N}_q'(t)} = \frac{\delta F_{q''}(t)}{\delta \hat{N}_q'(t)} = -k_B C_{q'q''}^{(-1)}(t), \tag{64} \]

that is, the second order functional derivatives of the IST-informational-entropy are the components of minus the inverse of the matrix of correlations \( C^{(-1)} \), with elements to be denoted by \( C_{q'q''}^{(-1)} \). Besides, the fluctuation of the IST-informational-entropy is given by

\[ \Delta^2 \hat{S}(t) = \sum_{q'q''} \frac{\delta S(t)}{\delta \hat{N}_q'(t)} \frac{\delta S(t)}{\delta \hat{N}_q''(t)} C_{q'q''}(t) = \sum_{q'q''} C_{q'q''}(t) F_{q'}(t) F_{q''}(t), \tag{65} \]
and that of the non-equilibrium thermodynamic variables $F_q(t)$ are

$$\Delta^2 F_q(t) = \sum_{q'q''} \frac{\delta F_q(t)}{\delta N_{q'}(t)} \frac{\delta F_q(t)}{\delta N_{q''}(t)} C_{q'q''}(t) = k_B \sum_{q'q''} C_{qq'}^{(-1)}(t) C_{qq''}^{(-1)}(t) = k_B^2 \delta C_{qq}(t), \quad (66)$$

therefore

$$\Delta^2 N_q(t) \Delta^2 F_q(t) = k_B^2 C_{qq}(t) \delta C_{qq}(t) = k_B^2 G_{qq}(t), \quad (67)$$

and then

$$[\Delta^2 N_q(t)]^{1/2} [\Delta^2 F_q(t)]^{1/2} = k_B [G_{qq}(t)]^{1/2}. \quad (69)$$

The quantities $C_{qq'}^{(-1)}$ are the matrix elements of the inverse of the matrix of correlations, and if the variables are uncorrelated $G_{qq}(t) = 1$. Equation (69) has the likeness of an uncertainty principle connecting the variables $N_q(t)$ and $F_q(t)$, which are thermodynamically conjugated in the sense of Eqs. (18) and (63), with Boltzmann constant being the atomic parameter playing a role resembling that of the quantum of action in mechanics. This leads to the possibility to relate the results of IST with the idea of complementarity between the microscopic and macroscopic descriptions of many-body systems advanced by Rosenfeld and Prigogine [52–55]; this is discussed elsewhere [56].

Care must be exercised in referring to fluctuations of the intensive variables $F_q$. In the statistical description fluctuations are associated to the specific variables $N_q$, but the $F_q$ are nonequilibrium thermodynamic intensive variables fixed by the average values of the $\hat{N}_q$, and so $\Delta^2 F_q$ is not a proper fluctuation of $F_q$ but a second order deviation interpreted as being a result of the fluctuations of the variables on which it depends, in a generalization of the usual results in statistical mechanics in equilibrium [57]. These brief considerations point to the desirability to develop a complete theory of fluctuations in the context of NESEF; one relevant application of it would be the study of the kinetics of transition between dissipative structures in complex systems, of which is presently available a phenomenological approach [58].

C. A Boltzmann-like relation: $\bar{S}(t) = k_B \ln W(t)$

According to the results of the previous subsection, quite similarly to the case of equilibrium it follows that the quotient between the root mean square of a given quantity and its average value is of the order of the reciprocal of the square root of the number of particles, that is

$$\frac{[\Delta^2 N_q(t)]^{1/2}}{N_q(t)} \sim N^{-1/2} \quad (70)$$

Consequently, again quite in analogy with the case of equilibrium, the number of states contributing for the quantity $N$ to have the given average value, is overwhelmingly enormous (a rigorous demonstration follows resorting to the method of the steepest descent [59]). Therefore,
we can write that

$$\phi(t) = \ln \text{Tr} \exp \left\{ - \sum_q F_q(t) \hat{N}_q \right\} \simeq \ln \left\{ W(t) \exp \left\{ - \sum_q F_q(t) \hat{N}_q \right\} \right\},$$

(71)

where

$$W(t) = \sum_{\tilde{n} \in \mathcal{M}(t)} 1 = \text{number of states in } \mathcal{M}(t),$$

(72)

where \(\tilde{n}\) is the set of quantum numbers which characterize the quantum-mechanical state of the system, and \(\mathcal{M}\) contains the set of states \(|\tilde{n}\rangle\) such that

$$\mathcal{M}(t) : \hat{N}_q(t) \leq \langle \tilde{n} | \hat{N}_q | \tilde{n} \rangle \leq \hat{N}_q(t) + \Delta \hat{N}_q(t),$$

(73)

where we have used the usual notations of bra and ket and matrix elements between those states. Hence we have that [cf. Eq. (51)]

$$\bar{S}(t) = k_B \bar{S}(t) = k_B \phi(t) + k_B \sum_q F_q(t) \hat{N}_q(t) \simeq k_B \ln W(t),$$

(74)

using Eq. (71), after disregarding the constant contribution from the thermal bath.

We recall that this is an approximate result, with an error of the order of the reciprocal of the square root of the number of degrees of freedom of the system, and therefore exact only in the thermodynamic limit.

Equation (74) represents the equivalent of Boltzmann expression for the thermodynamic entropy in terms of the logarithm of the number of complexions compatible with the macroscopic constraints imposed on the system.

Citing Jaynes, it is this property of the entropy - measuring our degree of information about the microstate, which is conveyed by data on the macroscopic thermodynamic variables - that made information theory such a powerful tool in showing us how to generalize Gibbs’ equilibrium ensembles to non-equilibrium ones. The generalization could never have been found by those who thought that entropy was, like energy, a physical property of the microstate [61]. Also following Jaynes, \(W(t)\) measures the degree of control of the experimenter over the microstate, when the only parameters the experimenter can manipulate are the usual macroscopic ones. At time \(t\), when a measurement is performed, the state is characterized by the set \(\{N_q(t)\}\), and the corresponding phase volume is \(W(t)\), containing all conceivable ways in which the final macrostate can be realized. But, since the experiment is to be reproducible, the region with volume \(W(t)\) should contain at least the phase points originating in the region of volume \(W(t_0)\), and then \(W(t) \geq W(t_0)\). Because phase volume is conserved in the micro-dynamical evolution, it is a fundamental requirement on any reproducible process that the phase volume \(W(t)\) compatible with the final state cannot be less than the phase volume \(W(t_0)\) which describes our ability to reproduce the initial state [61].
D. IST entropy and order parameter for magnons

In the cited “two-fluid model” the informational entropy, neglecting the constant part related to the bath, may be written as

\[
\bar{S}(t) = -n_1 \{N_1 \ln(N_1) - (N_1 + 1) \ln(N_1 + 1)\} - \\
- n_2 \{N_2 \ln(N_2) - (N_2 + 1) \ln(N_2 + 1)\},
\]

(75)

omitting to indicate the time dependence on the right for practical convenience.

The informational entropy in IST also satisfies a kind of generalized Clausius relation. In fact, consider the modification of the informational entropy as a consequence of the modification of external constraints imposed on the system. Let us call \( \lambda_\ell (\ell = 1, 2, \ldots, s) \) a set of parameters that characterize such constraints (e.g., the volume, external fields, etc.). Introducing infinitesimal modifications of them, say \( d\lambda_\ell \), the corresponding variation in the informational entropy, in the two-fluid model that was introduced, is given by

\[
d\bar{S}(t) = F_1(t) dN_1(t) + F_2(t) dN_2(t)
\]

(76)

where \( dN_{1,2}(t) \) are the nonexact differentials

\[
dN_{1,2}(t) = d\hat{N}_{1,2}(t) - \left\langle d\hat{N}_{1,2}|t\right\rangle,
\]

(77)

with \( \left\langle d\hat{N}_{1,2}|t\right\rangle = \text{Tr}\left\{d\hat{N}_{1,2}\hat{\rho}(t,0)\right\} \). In these expressions the nonexact differentials are the difference between the exact differentials

\[
dN_{1,2}(t) = d\text{Tr}\left\{\hat{N}_{1,2}\hat{\rho}(t,0)\right\} = \sum_{\ell=1}^{s} \frac{\partial N_{1,2}(t)}{\partial \lambda_\ell} d\lambda_\ell,
\]

(78)

and

\[
\left\langle d\hat{N}_{1,2}|t\right\rangle = \text{Tr}\left\{\frac{\partial \hat{N}_{1,2}(t)}{\partial \lambda_\ell} d\lambda_\ell \hat{\rho}(t,0)\right\},
\]

(79)

the latter being the average value of the change in the corresponding dynamical quantity due to the modification of the control parameters.

Equation (76) tells us that the non-equilibrium thermodynamic variables \( F_{1,2}(t) \) are integrating factors for the nonexact differentials \( d\hat{N}_{1,2}(t) \).

Using expression (75) it is possible to study the role of the Fröhlich contribution: changing the value of \( F \) (the coupling strength associated to Fröhlich contribution) in Eqs. (38), (39) and (40), we may virtually compare the informational entropy in systems with different Fröhlich coupling strengths.

The pumped system of magnons presented in Fig. 1 - where the magnon populations were numerically obtained from Eqs. (38) and (39) - has the informational entropy, obtained with the aid of Eq. (75), displayed as function of time in Fig. 4. In this figure we also show the time evolution of the informational entropy for the magnon populations obtained from Eqs. (38) and (39) with \( F = 0 \), i.e., a pumped magnon system with negligible Fröhlich contributions.

It can be noticed that the informational entropy values are lower when Fröhlich contribution is present, as it should as a result of having increasing ordering, that is, information increase. The same behavior occur in the case of the informational entropy of the steady states as function of the scaled rate of pumping where, as shown in Fig. 5, the presence of the Fröhlich contribution,
FIG. 4. Informational entropy as function of time associated with magnon populations from Eqs. (38) and (39) displayed in Fig. 1. Solid line represents the system with Fröhlich contribution, while the dashed line refers to a system in which the Fröhlich contribution is absent [F = 0 on Eqs. (38) and (39)]. Radiation pumping switched off at t = 1. We recall that the values of the different parameters are indicated in the caption of Fig. 1.

precisely in the region of the condensate, leads to a decrease of the informational entropy.

FIG. 5. Informational entropy of the steady states as function of the scaled rate of pumping I for systems with (solid) and without (dashed) Fröhlich contribution.

This decrease of informational entropy due to the Fröhlich contribution may be understood as some kind of increase in order and, to characterize this point, we introduce the order parameter

$$\Delta(F, I) = \frac{S_0^S(I) - S_F^S(I)}{S_0^S(I)} = 1 - \frac{S_F^S(I)}{S_0^S(I)},$$

where $S_0^S(I)$ and $S_F^S(I)$ are the steady-states informational entropies with and without Fröhlich contribution, that is

$$S_0^S(I) = f_1 \{(N_1^S + 1) \ln (N_1^S + 1) - N_1^S \ln (N_1^S)\} +
+ f_2 \{(N_2^S + 1) \ln (N_2^S + 1) - N_2^S \ln (N_2^S)\}$$

(81)
\[ S_0^S(I) = f_1 \left\{ \left( N_{S, F=0}^1 + 1 \right) \ln \left( N_{S, F=0}^1 + 1 \right) - N_{S, F=0}^1 \ln \left( N_{S, F=0}^1 \right) \right\} + \\
+ f_2 \left\{ \left( N_{S, F=0}^2 + 1 \right) \ln \left( N_{S, F=0}^2 + 1 \right) - N_{S, F=0}^2 \ln \left( N_{S, F=0}^2 \right) \right\}, \tag{82} \]

where the dependence of the steady-state populations on \( I \) has not been explicitly indicated. Fig. 6 presents the order parameter as function of the scaled rate of pumping which highlights this kind of complex order.

![Fig. 6. Order parameter of Eq. (80) as function of the scaled rate of pumping I.](image)

The role of the Fröhlich contribution may be evidenced through the numerical analysis of the order parameter as function of the Fröhlich contribution coupling strength when the rate of pumping is fixed. In Fig. 7 we present the mean steady-state populations \( N_{S, 1, 2} \) as function of \( F \) for fixed scaled rate of pumping \( I = 8 \times 10^{-4} \) and the corresponding informational entropy order parameter.

![Fig. 7. (a) Steady-state magnon populations as function of \( F \). (b) The related order parameter.](image)

As can be seen in the Fig. 7(a), the magnon steady-state populations \( N_{S, 1, 2} \) decrease as the nonlinear Fröhlich contribution coupling strength increases (notably the mean population associated with high frequencies magnons \( N_{S, 2}^2 \)). This complex behavior of the steady-state populations may be understood considering that: (i) the Fröhlich contribution leads to the formation of the condensate in which the magnons with lower frequency are overpopulated at the expense
of the higher in frequency populations; (ii) since the substantial decrease of \(N_S^2\) relative to \(N_S^1\) diminishes the absorbance of the material [because of the positive feedback effect of the parallel pumping, see Eq. (39a)] the net flux of absorbed energy is lower than in the case without Fröhlich interaction, justifying the global fall of the mean population. The order parameter behavior, shown in Fig. 7(b), corroborates the idea that Fröhlich contribution enhances the complex order mentioned before.

E. IST entropy production

We analyze the informational-entropy production and, using Eq. (51) [paying attention to the logarithm of the partition functions in Eq. (52)], it can be shown that it is given by

\[
\bar{\sigma}(t) = \frac{d}{dt} \bar{S}(t) = \beta_0 \frac{dE_B(t)}{dt} + \sum_q F_q(t) \frac{dN_q(t)}{dt},
\]

and then, taking into account Eqs. (27) and (34), it can be rewritten in terms of two contributions

\[
\bar{\sigma}(t) = \bar{\sigma}_i(t) + \bar{\sigma}_e(t),
\]

consisting of the so-called internal one, \(\bar{\sigma}_i(t)\), which results from internal interactions in the system, and the external one, \(\bar{\sigma}_e(t)\), related to the interactions with the surroundings, in this case with the source and the thermal reservoir. They are given by

\[
\bar{\sigma}_i(t) = \sum_q \left\{ F_q(t) \left[ \Omega_q(t) + L_q(t) + \varphi_q(t) + \Lambda_q(t) \right] - \beta_0 \hbar \omega_q \left[ \Omega_q(t) + L_q(t) + \varphi_q(t) \right] \right\},
\]

\[
\bar{\sigma}_e(t) = \sum_q \left\{ F_q(t) \Omega_q(t) + \beta_0 \frac{J_{(2)}}{T^2} \right\},
\]

or, using Eq. (36),

\[
\bar{\sigma}_e(t) = \sum_q \left\{ F_q(t) \Omega_q(t) + \beta_0 \hbar \omega_q \left[ \Omega_q(t) + L_q(t) + \varphi_q(t) \right] \right\}.
\]
In the two-fluid model the informational-entropy production is thus given by

\[ \dot{\sigma}_1(t) = \sum_q \left\{ [F_q(t) - \beta_0 \hbar \omega_q] \left[ \mathfrak{R}_q(t) + L_q(t) + \mathfrak{F}_q(t) \mathfrak{M}_q(t) \right] + F_q(t) \mathfrak{M}_q(t) \right\} \approx \left( \ln \left( \frac{N_1 + 1}{N_1} \right) - \beta_0 \hbar \omega_1 \right) \sum_{q \in R_1} \left[ \mathfrak{R}_q(t) + L_q(t) + \mathfrak{F}_q(t) \mathfrak{M}_q(t) \right] + \\
+ \left\{ \ln \left( \frac{N_2 + 1}{N_2} \right) - \beta_0 \hbar \omega_2 \right\} \sum_{q \in R_2} \left[ \mathfrak{R}_q(t) + L_q(t) + \mathfrak{F}_q(t) \mathfrak{M}_q(t) \right] + \\
+ \ln \left( \frac{N_1 + 1}{N_1} \right) \sum_{q \in R_1} \mathfrak{M}_q(t) + \ln \left( \frac{N_2 + 1}{N_2} \right) \sum_{q \in R_2} \mathfrak{M}_q(t) = \\
= - \frac{\eta}{\tau} \left\{ \ln \left( \frac{N_1 + 1}{N_1} \right) - \beta_0 \hbar \omega_1 \right\} \left\{ D \mathcal{N}_1 (\mathcal{N}_1 - \mathcal{N}_1^{(0)}) + f_1 \left[ \mathcal{N}_1 - \mathcal{N}_1^{(0)} \right] \right\} + \\
+ \frac{\eta}{\tau} \left\{ \ln \left( \frac{N_1 + 1}{N_1} \right) - \beta_0 \hbar \omega_1 \right\} F \{ \mathcal{N}_1, \mathcal{N}_2 + (\nu + 1) \mathcal{N}_2 - \nu \mathcal{N}_1 \} - \\
- \frac{\eta}{\tau} \left\{ \ln \left( \frac{N_2 + 1}{N_2} \right) - \beta_0 \hbar \omega_2 \right\} \left\{ D \mathcal{N}_2 (\mathcal{N}_2 - \mathcal{N}_2^{(0)}) + f_2 \left[ \mathcal{N}_2 - \mathcal{N}_2^{(0)} \right] \right\} - \\
- \frac{\eta}{\tau} \left\{ \ln \left( \frac{N_2 + 1}{N_2} \right) - \beta_0 \hbar \omega_2 \right\} F \{ \mathcal{N}_1, \mathcal{N}_2 + (\nu + 1) \mathcal{N}_2 - \nu \mathcal{N}_1 \} + \\
+ \frac{\eta}{\tau} \left\{ \ln \left( \frac{N_2 + 1}{N_2} \right) - \ln \left( \frac{N_1 + 1}{N_1} \right) \right\} M \{ \mathcal{N}_1 (\mathcal{N}_1 + 1) + \mathcal{N}_2 (\mathcal{N}_2 + 1) \} \times \\
\times (\mathcal{N}_1 \mathcal{N}_2^{(0)} - \mathcal{N}_2), \right. 
\]
and
\[
\bar{\sigma}_e(t) = \\
= \sum_q \{ F_q(t) \bar{\sigma}_q(t) + \beta_0 \hbar \omega_q [ R_q(t) + L_q(t) + \bar{\delta}_q(t)] \} \approx \\
\approx \ln \left( \frac{N_1 + 1}{N_1} \right) \sum_{q \in R_1} \bar{\sigma}_q(t) + \ln \left( \frac{N_2 + 1}{N_2} \right) \sum_{q \in R_2} \bar{\sigma}_q(t) + \\
+ \beta_0 \hbar \omega_1 \sum_{q \in R_1} [ R_q(t) + L_q(t) + \bar{\delta}_q(t)] + \\
+ \beta_0 \hbar \omega_2 \sum_{q \in R_2} [ R_q(t) + L_q(t) + \bar{\delta}_q(t)] = \\
= \frac{n}{\tau} \ln \left( \frac{N_2 + 1}{N_2} \right) I (1 + 2N_2) + \\
+ \frac{n}{\tau} \beta_0 \hbar \omega_1 \left\{ -D N_1 (N_1 - N_1^{(0)}) - f_1 \left[ N_1 - N_1^{(0)} \right] + \\
+ F [ N_1 N_2 + (\bar{\nu} + 1) N_2 - \nu N_1 ] \right\} + \\
+ \frac{n}{\tau} \beta_0 \hbar \omega_2 \left\{ -D N_2 (N_2 - N_2^{(0)}) - f_2 \left[ N_2 - N_2^{(0)} \right] - \\
- F [ N_1 N_2 + (\bar{\nu} + 1) N_2 - \nu N_1 ] \right\}, \tag{90}
\]
where we used that
\[
\ln \left( \frac{N_{1,2}^{(0)} + 1}{N_{1,2}^{(0)}} \right) = \beta_0 \hbar \omega_{1,2}, \tag{91}
\]
with $N_{1,2}^{(0)}$ being the distribution in equilibrium.

![Figure 8](image-url)

**FIG. 8.** Informational-entropy production of the system of magnons as function of time (associated with Figs. 1 and 4). On the left it can be observed the internal, external and total entropy production (and we call attention to the expected non-negative values of the internal entropy production); On the right only the total entropy production is shown. We recall that the pumping source is switched off at $\bar{t} = 1$.

In Fig. 8 we show the informational-entropy production of the system of magnons evolving
in time (i.e., the entropy production associated with the evolution described in Fig. 1). We can observe that, although the internal entropy production is strictly non-negative - as it should -, the total entropy production may have positive and negative values. This is related, in informational non-equilibrium statistical thermodynamics, with the generalized $H$-theorem in the sense of Jancel [62], which we have called weak principle of increasing of informational entropy, namely, given the informational statistical entropy $\tilde{S}(t)$ of Eq. (51) and the informational entropy production $\sigma(t)$ of Eq. (83), the principle tells us that
\[
\Delta \tilde{S}(t) = \tilde{S}(t) - \tilde{S}(t_0) = \int_{-\infty}^{t} dt' \sigma(t') \geq 0.
\]

(92)

Equation (92) does not prove that $\sigma(t)$ is a monotonically increasing function of time, as required by phenomenological irreversible thermodynamic theories. We have only proved the weak condition that as the system evolves it is predominantly definite positive. We stress the fact that this result is a consequence of the presence of the irreversible part of $\hat{\rho}(t)$ not contained in $\hat{\rho}(t)$, which is then, as stated previously, the part that accounts for - in the description of the macroscopic state of the system - the processes which generate dissipation. Furthermore, the informational entropy with the evolution property of Eq. (92) is the coarse-grained entropy of Eq. (48), the coarse-graining being performed by the action of the projection operator $P_\varepsilon(t)$ of Eqs. (49) and (50): This projection operation extracts from the Gibbs entropy the contribution associated to the constraints [cf. set (7)] imposed on the system, by projecting it onto the subspace spanned by the basic dynamical quantities (see also [51]). Hence, the informational entropy thus defined depends on the choice of the basic set of macroscopic variables, whose completeness in a purely thermodynamic sense cannot be indubitably asserted. We restate that in each particular problem under consideration the information lost as a result of the particular truncation of the set of basic variables must be carefully evaluated [63, 64]. Retaking the question of the signal of $\sigma(t)$, we conjecture that it is always non-negative, since it can not be intuitively understood how information can be gained in some time intervals along the irreversible evolution of the system. However, this is expected to be valid as long as we are using an, in a sense, complete description of the system. Once a truncation procedure is introduced [58] the local density of informational entropy production is no longer monotonously increasing in time; this has been illustrated by Criado-Sancho and Llebot [62] in the realm of Extended Irreversible Thermodynamics, and in IST in [66]. The reason is, as pointed out by Balian et al. [51] that the truncation procedure introduces some kind of additional (spurious) information at the step when the said truncation is imposed.

Another important result, shown in Fig. 9, is the informational-entropy production for the steady states (see Fig. 2).
F. The Evolution Criterion

The change in time of IST-entropy production can be separated into two parts, namely

$$\frac{d}{dt} \bar{\sigma}(t) = \frac{dQ}{dt} \sigma(t) + \frac{dF}{dt} \bar{\sigma}(t),$$

(93)

where

$$\frac{dQ}{dt} \sigma(t) = \sum_q F_q(t) \frac{d^2 N_q(t)}{dt^2},$$

(94)

that is, the part that accounts for the change in time of $N_q(t)$, and

$$\frac{dF}{dt} \bar{\sigma}(t) = \sum_q \frac{dF_q(t)}{dt} \frac{dN_q(t)}{dt},$$

(95)

accounting for the part of change in time of the non-equilibrium thermodynamics variables $F_q(t)$.

Recalling that $F_q(t)$ may be expressed in terms of the populations [cf. Eq. (21)], we have that

$$\frac{dF_q(t)}{dt} = \frac{d}{dt} \ln \left\{ \frac{N_q(t) + 1}{N_q(t)} \right\} = \left[ \frac{1}{N_q(t) + 1} - \frac{1}{N_q(t)} \right] \frac{dN_q(t)}{dt} = -\frac{1}{(N_q + 1)N_q} \frac{dN_q(t)}{dt},$$

(96)

and therefore

$$\frac{dF}{dt} \bar{\sigma}(t) = -\sum_q \frac{1}{(N_q + 1)N_q} \left( \frac{dN_q}{dt} \right)^2 \leq 0.$$  (97)

This inequality verifies for this system the generalization of Glansdorff–Prigogine’s thermodynamic criterion of evolution\[47, 48, 67\]. That is, along the trajectory of the macrostate of the system in the thermodynamic (or Gibbs) space of states, the quantity of Eq. (93) is always non-
positive, a quantity which in classical Onsagerian thermodynamics is the product of the change in time of the thermodynamic forces times the fluxes of matter and energy.

G. The (In)stability Criterion

Within the above discussed framework of a non-equilibrium thermodynamics of the Fröhlich-Bose-Einstein condensation of magnons, we may analyze again the stability of the steady-states populations $N_q$. Considering arbitrary small deviations, say $\epsilon \eta_q(t)$, from the steady state, we may expand the informational entropy in the form

$$S \left( \{ N_q \} \right) = S \left( \{ N_q^S \} \right) = S \left( \{ N_q^S \} \right) + \delta S + \delta^2 S + \ldots,$$

with

$$\delta^n S = \frac{\partial^n S}{\partial \epsilon^n} \bigg|_{\epsilon=0} \frac{\epsilon^n}{n!}.$$  

Since

$$\frac{\partial^2 S}{\partial \epsilon^2} = -\sum_q \frac{\eta_q^2(t)}{[N_q^S + \epsilon \eta_q(t) + 1]} \left[ \frac{\Delta N_q(t)}{[N_q^S + \epsilon \eta_q(t) + 1]} \right],$$

we have that the second variation of the entropy is

$$\delta^2 S = -\sum_q \frac{\epsilon^2 \eta_q^2(t)}{[N_q^S + 1]} \Delta N_q(t) = -\sum_q \frac{[\Delta N_q(t)]^2}{(N_q^S + 1) N_q^S} \leq 0,$$  

where $\Delta N_q(t)$ represents the value of the imposed arbitrary deviation from the steady state and the non-positiveness of Eq. (101) is a manifestation of the convexity of the maximized informational entropy. Differentiation in time of Eq. (101) introduces the quantity called excess of entropy production function, namely

$$\delta^2 \sigma(t) = \frac{1}{2} \frac{d}{dt} \delta^2 S(t) = -\sum_q \frac{\Delta N_q(t)}{(N_q^S + 1) N_q^S} \frac{d}{dt} \Delta N_q(t),$$

which, in the two fluid model has the following form

$$\delta^2 \sigma(I, t) = -\frac{n_1}{(N_1^S(I) + 1) N_1^S(I)} \frac{d}{dt} \Delta N_1(I, t) - \frac{n_2}{(N_2^S(I) + 1) N_2^S(I)} \frac{d}{dt} \Delta N_2(I, t).$$

According to Glansdorff-Prigogine (in)stability criterion[47, 48, 67], if

$$\frac{1}{2} \delta^2 S(t) \delta^2 \sigma(t) \leq 0,$$

then the steady-state is stable, and this is so once $\Delta N_{1,2} \frac{dN_{1,2}}{dt} \leq 0$, as it follows from solving the evolution equations, Eqs. (38) and (39). Then $\delta^2 \sigma \geq 0$ and, since $\delta^2 S \leq 0$, Eq. (104) is verified. Hence, for the given constraints the reference steady state is stable for all fluctuations.
compatible with the equations of evolution.

The stability has been derived in relation to homogeneous fluctuations, but instability may emerge due to induced inhomogeneous states. Some experimental observations may be pointing in that direction, and the question is under consideration.

Using linear stability analysis it was previously shown the stability of the resulting steady state (cf. Fig. 3). Such stability has been here rederived using Glansdorff-Prigogine analysis which involves physical considerations instead of only mathematical ones, with Glansdorff-Prigogine’s excess entropy production function being a Lyapunov function for this system.

IV. CONCLUDING REMARKS

We have considered the non-equilibrium statistical thermodynamics of the Bose-Einstein condensation of magnons excited under the action of radio-frequency radiation pumping. It has better referred to as Fröhlich-Bose-Einstein condensation, once, as noticed, the phenomenon which is possible to emerge in systems of bosons, was originally evidenced by Herbert Fröhlich [13, 14].

It constitutes an example of complexity in which, after a certain threshold in the value of the intensity of the pumping source has been attained, there follows that the energy pumped on the system is transferred from modes higher in frequency to those lower in frequency in a cascade-down-type process. The modes of lowest energy are then largely populated at the expense of the other modes with higher frequencies. The emergence of the phenomenon is driven by a nonlinear interaction (involving two magnons and one phonon), which is in competition (kind of “tug of war”) with the magnon-magnon interaction. As a result, it can be evidenced the existence of three regimes depending on the intensity of the pumping source: at low intensities there follows a simple linear behavior, as it should according to Prigogine’s principle of minimum local-equilibrium entropy; followed, as the pumping intensity is increased, by a regime of formation of the Fröhlich-Bose-Einstein condensate; and finally at a further threshold of pumping intensity there follows a regime of simple thermal distribution (when the magnon-magnon interaction overcomes the non-linear interaction that drives the emergence of the condensate).

We have introduced a “two-fluid model” and the coefficients present in the kinetic equations were evaluated and finally adjusted using comparison with the experimental results in YIG.

In the non-equilibrium thermodynamic analysis we have considered several important characteristics. First we have derived the so-called informational entropy for the system of magnons (Section III A). In Section III B are considered the fluctuations of the population operator and the associated Maxwell relations. Furthermore, we have shown the derivation of a generalized $\mathcal{H}$-theorem, with the derivation of a Boltzmann-like relation for the non-equilibrium statistical entropy, that is, given by the logarithm of the number of complexions compatible with the non-equilibrium macroscopic constraints imposed on the system (Section III C).

In Section III D we have specified the magnon informational entropy for the “two-fluid model” and shown that the system obeys a kind of generalized Clausius relations. Then, in terms of entropy production, we introduced an order parameter. Through this order parameter, the informational entropy is shown to be smaller when the nonlinear interaction responsible for the onset of the NEFBEC predominates, thus evidencing the increase of order due to the Fröhlich interaction.

It has also been calculated the informational-entropy production function (Section III E), characterizing the contributions of the internal and external informational entropy production, with the former having, as it should, non-negative values characterizing dissipation, while the external one is negative as a result of the pumping on the system.

Finally, it has been verified that Glansdorff-Prigogine evolution criterion is satisfied (Section III F), and from the generalization of Glansdorff-Prigogine (in)stability principle we have shown that the non-equilibrium thermodynamic state of system is stable under any condition (Section
We stress that such instability has been derived in relation to other possible homogeneous state, but instability against the onset of a spatially ordered state cannot be ruled out, and is being under consideration.

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