Nonlinear relaxation in nonequilibrium oscillators: Bose–Einstein-like condensation in a dissipative structure

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Abstract

We consider a system of externally pumped nonequilibrium harmonic oscillators (vibrational modes) interacting through anharmonic effects with a system of harmonic oscillators with lower-lying frequencies (acoustic-like vibrations) composing a thermal bath for the former. We derive the equations of evolution for the population of the vibrational modes, introducing high order nonlinear relaxation processes. It is shown that complex behavior arises in this system, namely, after a certain critical intensity of the pumping source is achieved, the populations of the modes lowest in frequency increase enormously at the expense of the excitations of the other modes. In this way there follows what can be termed a Bose–Einstein-like condensation, not in a phase in equilibrium, but in a nonequilibrium dissipative structure. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

Nonlinearity in physical, chemical and biological systems is the source of new and unexpected complex behavior. Complexity manifests itself particularly in two kinds of situations related to dynamical systems: One is chaotic behavior in mechanical systems, the other is the case of open systems driven far from equilibrium by intense external sources, where it is possible to find the emergence of ordered patterns at the macroscopic scale [1,2]; the present paper belongs to this area.

The concept that many-body systems sufficiently far from equilibrium and governed by nonlinear kinetic laws may display self-organized ordered structures at the macroscopic level, as observed in many cases, has been brought under unifying approaches such as dissipative structures [3–8], synergetics [9], and macro-concepts [10].

We deal in this paper with a system of harmonic oscillators (as vibrational modes) driven farther and farther away from equilibrium by an external source that pumps energy on the system, while it is in contact with an external thermal bath consisting of a system of vibrational modes. Harmonic oscillators play an important role in the description of physical systems: We can mention their fundamental role in the description of lattice vibrations in solids (phonons in the quantized form), as well as in the description of the dynamics of excitations like plasmons, polaritons, plasmaritons, magnetoelectronic waves, etc. They are also present in the description of biomaterials as normal mode excitations, for example in long chains of macromolecules coupled by peptide groups sustaining dipolar oscillations. In these materials (solid state or

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biological) are usually present high frequency (infrared region) polar modes and low frequency acoustic modes, both interacting through anharmonic effects, like in the model presented in Section 4.

For the study of the dissipative systems of oscillators described at the beginning of the preceding paragraph, we resort to a seemingly powerful, and also elegant and concise, mechanical–statistical formalism, namely, the Nonequilibrium Statistical Operator Method (NESOM). It has been the object of several approaches which, it can be shown [11–18], can be placed within the context of a unifying variational procedure based on Jaynes’ Predictive Statistical Mechanics [19–22]. NESOM allows for the construction of a nonlinear quantum transport theory – a far-reaching generalization of the Chapman–Enskog’s and also Mori’s methods, which describes the evolution of the system at the macroscopic level in arbitrary nonequilibrium situations. Among the different approaches to NESOM we resort here to the use of Zubarev’s approach [11–18] and the nonlinear quantum kinetic theory that the formalism provides [23–26].

We derive the equations of evolution for the population of the vibrational modes characterized by a given frequency dispersion relation. We take a periodic distribution of the oscillating centers, and then the wave vector in the dispersion relation which runs over a Brillouin zone, like, for example, polar modes in solids [27] or dipolar vibrational centers in biopolymers [28]. The bath, composed of a subsystem of acoustic-like vibrations is assumed to remain, through a very effective thermal contact, in equilibrium with an ideal reservoir at temperature $T_0$.

In the next sections, after the derivation and discussion of the equations of evolution, provided by the above mentioned NESOM-based kinetic theory, we proceed to a numerical approximate calculation, looking for the values of the populations of the vibrational modes in the steady state in terms of the intensity of the external energy pumping source. Our results clearly evidence a complex behavior consisting of a phenomenon conjectured by Fröhlich [29–31] (we call it Fröhlich effect) namely, that after a critical value in the intensity of the external pumping source is achieved, in a cascading-down process the vibrational modes transfer large part of the energy they are receiving to a set of vibrational modes with the lowest frequencies (those of large wave vectors at the zone boundary in our model) in a way akin to a Bose–Einstein condensation, not for phases in equilibrium, but in nonequilibrium dissipative structures. It should be reinforced that this notable and unexpected phenomenon is a result of nonlinearities in the equations of evolution that describe the macroscopic state of the system as dissipative processes develop in it.

2. Equations of evolution for the vibrational modes

We consider a periodic array of harmonic oscillators which have associated vibrational modes, consisting of a high frequency branch with frequency dispersion relation $\omega_k$, and a low frequency branch (acoustic-like branch) with frequency dispersion relation $\Omega_k$. The wave-vector $k$ runs over the reciprocal space Brillouin zone. Further, an external source continuously pumps energy on the upper branch of oscillators, while the acoustic-like branch is taken as a thermal bath constantly kept at temperature $T_0$ through a good thermal contact with a reservoir.

We write for the system Hamiltonian

$$H = H_0 + H',$$  

(1)

according to the requirement of the NESOM [11–18], where

$$H_0 = \sum_k \hbar \omega_k \left( a_k^+ a_k + \frac{1}{2} \right) + \sum_k \hbar \Omega_k \left( b_k^+ b_k + \frac{1}{2} \right)$$  

(2)

and

$$H' = H_1' + H_{11}' + H_{12}' + H_{21}' + H_{22}'$$  

(3)

with

$$H_i' = \sum_k \phi_k a_k^+ + \mathcal{H},$$  

(4)
\begin{align}
H_{11}' &= \sum_{k,q} V_{kq} a_k^q b_{k-q}^+ + \text{HC}, \\
H_{12}' &= \sum_{k,q} V_{kq} a_k^q b_{k+q}^+ + \text{HC}, \\
H_{21}' &= \sum_{k,q} V_{kq} a_k^q b_{k-q} + \text{HC}, \\
H_{22}' &= \sum_{k,q} V_{kq} a_k^q a_{k+q}^+ + \text{HC}. 
\end{align}

In these equations \(a(a^+)\) and \(b(b^+)\) are the annihilation (creation) operators of the upper branch vibrational modes and of the vibrational modes of the thermal bath, respectively. The two terms in \(H_0\) are the Hamiltonians of the free subsystems, \(H_i'\) accounts for the interaction between the pumping source and the vibrational modes, with \(\varphi\) and \(\varphi^+\) being annihilation and creation operators for the excitations in the source, also incorporating the coupling strength. The other terms, namely \(H_{ij}'\) with \(i,j = 1,2\), are the anharmonic interactions between both types of vibrations that contribute to the equations of evolution. Momentum conservation has been taken into account, and we noticed that this is a truncated Hamiltonian in the so-called Rotating Wave Approximation [32], a satisfactory one in the present case. The sums over the reciprocal space vectors \(k\) and \(q\) run, we recall, over the Brillouin zone.

To deal with this system in NESOM, the first step is to define the basic set of variables which provide the description of its macroscopic state [11–18]. Considering the problem in hand, that is, arbitrarily excited modes in the upper branch of the vibrational modes of the system, and the lower branch of acoustic-like vibrations assumed to be constantly in equilibrium with a reservoir at temperature \(T_0\), we choose for the basic set of macrovariables (1) the population of the vibrational modes,

\[ v_k(t) = \text{Tr}\{a_k^+ a_k \rho_a(t)\}, \tag{6} \]

and (2) the energy of the thermal bath,

\[ E_b(t) = \text{Tr}\{H_b \rho_a(t)\} = \text{Tr}\left\{\sum_k \hbar \Omega_k \left( b_k^+ b_k + \frac{1}{2}\right) \rho_a(t) \right\}, \tag{7} \]

where \(H_b\) is the Hamiltonian of free thermal bath, \(\rho_a(t)\) is Zubarev’s nonequilibrium statistical operator for this case, when the auxiliary operator is then given by [11–18]

\[ \tilde{\rho}(t,0) = \exp\left\{ -\phi(t) - \sum_k F_k(t) a_k^+ a_k - \beta_0 H_b \right\}, \tag{8} \]

where \(F_k(t)\) is the nonequilibrium thermodynamic parameter conjugated to the dynamical variable population of the vibrational modes, and \(\phi(t)\) ensures the normalization of the statistical operator. Moreover, \(\beta_0 = 1/k_b T_0\) where \(k_b\) is as usual Boltzmann constant and \(T_0\) is the constant temperature of the thermal bath, which is a consequence of the hypothesis that the acoustic modes are constantly in thermal equilibrium with the reservoir at temperature \(T_0\). This implies that the energy of the thermal bath, as given by Eq. (7), increases as a result of the energy pumped by the excited (out of equilibrium) system, but decreases as the energy pumped in excess of equilibrium is transferred to the external reservoir, and both processes compensate each other. That is, considering that the equation of evolution for \(E_b(t)\) is

\[ \frac{d}{dt} E_b(t) = \dot{E}_b(t)|_{\text{pump}} + \dot{E}_b(t)|_{\text{relax}} \tag{9} \]

with the two terms on the right accounting for the above-said processes, and we recall that we have assumed that a very good thermal contact is present between bath and reservoir such that \(\frac{dE_b(t)}{dt} \approx 0\), once as noticed, the thermal bath and thermal reservoir are constantly in mutual equilibrium at the temperature \(T_0\) of the latter. Hence,
\[ E_B = E_B^0 = \sum_k \hbar \Omega_k \left( \zeta_k + \frac{1}{2} \right), \]

where
\[ \zeta_k = \left[ \exp \{ \beta \hbar \Omega_k \} - 1 \right]^{-1} \]
is the population in equilibrium at temperature \( T_0 \).

Consequently, we are left with the task of evaluating the equation of evolution for the population of vibrational modes, what we do resorting to the NESOM-based kinetic theory [23–26], to obtain that
\[
\frac{d}{dt} v_k = I_k(\omega_k) - \frac{1}{\tau_k} [v_k(t) - v_k^0] - \sum_q M^{(1)}_{kq} \left\{ v_q(t) v_q(t) e^{\hbar (\omega_k + \omega_q)} - [1 + v_q(t)] [1 + v_q(t)] \right\} \\
+ \sum_q M^{(2)}_{kq} \left\{ v_q(t) [1 + v_q(t)] e^{\hbar (\omega_k - \omega_q)} - v_k(t) [1 + v_k(t)] \right\} + \sum_q A_{kq} \left\{ v_q(t) [1 + v_k(t)] \right\}
\]

where the different coefficients \( I, \tau, M, \) and \( A \) are listed in Appendix A, and \( v_k^0 \) is the population of the polar modes in equilibrium at temperature \( T_0. \) As already noticed, Eq. (12) follows from the application of the generalized nonlinear quantum transport theory derived in the framework of the NESOM [23,32], applied in this case to the quantity \( v_k(t) \) of Eq. (6). We recall that the fourth-order approximation we use is composed of a number of contributions, as discussed in Refs. [23–26]. However, for this particular problem, symmetry properties make null several terms, and those surviving in Eq. (12) are those that correspond:

(a) To an equivalent of the Golden Rule of quantum mechanics averaged over the nonequilibrium ensemble – they are terms with coefficients \( \tau^{-1}, M^{(1)} \) and \( M^{(2)} \), but it must be noticed that the two with \( M^{(1)} \) vanish because they correspond to processes that are not allowed because energy conservation in the scattering event is not possible (we recall that the frequency of the phonons of the bath is lower than that of the vibrations under consideration), and \( M^{(2)} \) gives a negligible contribution in this case of a very narrow frequency-dispersion relation \( \omega_q \); (b) To the terms, those with coefficient \( A \), that are the fourth-order contribution in Born’s perturbation series averaged over the nonequilibrium ensemble.

The equation of evolution, Eq. (12), for the population of the \( k \)-mode is composed of several contributions: The first is the one associated to the pumping effects (from the external source) which leads the system further and further away from equilibrium as \( I \) (the intensity of the source) is increased. The second contribution accounts for relaxation of the population in excess of equilibrium, created by the source, to the thermal bath thus diminishing the value of the population. The third term (the one with \( M^{(1)} \)) and the fourth term (the one with \( M^{(2)} \)), as noticed, do not contribute, and finally, the fifth term (the one with \( A \)) contains a nonlinear contribution, expressed by
\[
A_{kq} v_k(t) v_q(t) [1 - e^{\hbar A_{kq}}],
\]

which can produce either a relaxation or an excitation effect depending on the sign of the difference \( A_{kq} = \omega_k - \omega_q \). A given mode \( k \) tends to increase its population at the expense of the other modes \( q \) if it is verified that \( \omega_q > \omega_k \). This clearly implies that the energy pumped by the external source on the different modes would tend to be transferred to the modes with the lowest frequencies in a cascading-like process.

When a constant external source acts continuously, after a transient time has elapsed a steady state, i.e., \( dv/dt = 0 \), must follow (the steady state in the absence of the source (\( I = 0 \)) is the equilibrium state). Let us next look for the characteristics of this steady state. First, we notice that
\[
v_k(t) = \text{Tr} \{ a_k^+ a_k \rho(t, 0) \} = \left[ e^{\xi_k(t)} - 1 \right]^{-1},
\]

which follows in a straightforward way by simply calculating the trace operation with the distribution of Eq. (8). In equilibrium, \( F_k = \beta \hbar \omega_k \) and Eq. (14) is the Planck distribution of the populations. Consider now the nonequilibrium steady-state situation when the now time-independent nonequilibrium intensive variable \( F_k \) can be redefined as
\[ F_k = \beta [\hbar \omega_k - \mu_k]. \]  

(15)

Hence, replacing Eq. (15) into Eq. (14) we find that

\[ v_k = \left[ \exp \left( \beta (\hbar \omega_k - \mu_k) \right) \right]^{-1}, \]  

(16)

where the quantity \( \mu_k \) is given in Appendix B.

Eq. (16) is an interesting alternative form of Eq. (14) in the steady state. It resembles a Bose–Einstein distribution with temperature \( T_0 \) and a quasi-chemical potential \( \mu_k \) for each mode. We call attention to the fact that this quasi-chemical potential is a complicated functional of the population of all the modes (see Appendix B). Moreover, the quasi-chemical potential per mode goes to zero in the limit of a vanishing pumping source, and then Eq. (16) becomes the Planckian distribution in equilibrium. But in the presence of a pumping source, \( \mu_k \) is positive and growing with the increasing intensity of the source. This is an indication that it may expected that the most favored mode – the one with the lowest frequency, may be lead to a situation when, for a sufficiently high intensity of the source, its quasi-chemical potential may approach its frequency and a Bose-like condensation would follow. We analyze this possibility Section 3 on the basis of a simplified model. As a final word in this section we note that the concept of Bose–Einstein distributions with nonzero quasi-chemical potential in the nonequilibrium populations of bosons, which are otherwise Planck distributions in equilibrium, have been used by Landsberg for the populations of photons in the case of a steady state between radiation and an electron-hole plasma in semiconductors [33], and by Fröhlich for the characterization of the population of polar wave excitations in biophysical systems [29–31].

Moreover, an alternative form for the Lagrange multiplier is

\[ F_k = \hbar \omega_k / k_\beta T^*, \]  

(17)

what defines a so-called quasi-temperature, a concept widely used in the physics of semiconductors under highly excited conditions of excitations [34–36]. Both concepts, of quasi-chemical potential and quasi-temperature in the thermodynamics of dissipative systems are discussed in Ref. [37,38].

3. Numerical solutions for a model system

The equations of evolution for different modes are a set of nonlinear integro-differential equations that couple all the modes. To perform numerical solutions we resort to a simplified model: Noting the already referred-to effect that the modes at the lowest frequencies receive the energy pumped on all the other modes higher in frequency through the last term in Eq. (12), or, more precisely, the contribution indicated in Eq. (13), in a cascading-down process, we introduce a two-fluid-like model, similar to the case of phase transitions leading to superfluidity and superconductivity. One is the ‘normal fluid’, in this case consisting of the set of modes higher in frequency that are excited by the energy-pumping external source, and the ‘superfluid’, or the condensate, consisting of the set of modes lower in frequency that draw energy from the others. We indicate by \( g_n \) and \( g_c \) the number of modes that compose, respectively, those two sets of modes, and \( \omega_n \) and \( \omega_c \) (with \( \omega_n > \omega_c \)) designating the characteristic frequencies for each set assuming that the band of frequencies \( \omega_k \) has a narrow width, what is usually the case in real situations.

Moreover, we use a Debye model for the vibrational modes of the thermal bath, namely, we take \( \Omega_q = s|q| \), where \( s \) is the group velocity of propagation. Also, we take the matrix elements \( |V_{0k}|^2 \) out of the integral sign (the sum over \( q \) in the spirit of the mean-value theorem of calculus, and we write \( V(k) \) for this value, which is a posteriori evaluated through the use of Eq. (A.1) in Appendix A to obtain

\[ |V(k)|^2 = 2\pi \hbar \frac{\beta^2 \omega_0}{\gamma \tau_k}, \]

(18)

In words, we have related this unknown coefficient to a phenomenological parameter, namely, the relaxation time \( \tau_k \), which can be experimentally determined from Raman bandwidths. Moreover, \( \omega_0 \), we recall, is the distribution in equilibrium, and \( \gamma \) is the volume of the system.
The two coupled equations for the populations in the steady state, namely, for the ‘normal’ modes and the modes in the ‘condensate’, designated $\tilde{v}_n$ and $\tilde{v}_c$ respectively, are

$$I_n - \gamma_n^{-1}(v_n - v_n^0) - g_n R_{nc} = 0,$$

(19a)

$$I_c - \gamma_c^{-1}(\tilde{v}_c - \tilde{v}_c^0) + g_c R_{nc} = 0,$$

(19b)

where

$$R_{nc} = A_{nc} \left\{ \tilde{v}_n \left(1 + \tilde{v}_c\right) e^{\Delta_n} - \tilde{v}_c \left(1 + \tilde{v}_n\right) \right\}.$$  

(20)

The coefficients $g_m$ measuring the number of modes comprising the normal part and the condensate are proportional to the extension of the system, and we take for them the expressions

$$g_n = \beta_n \frac{\gamma}{2\pi^3} \frac{4}{3} \pi Q^3_B,$$

(21a)

$$g_c = \beta_c \frac{\gamma}{2\pi^3} \frac{4}{3} \pi Q^3_B,$$

(21b)

where $\beta_n$ and $\beta_c$ (with both smaller than one, and $\beta_n + \beta_c = 1$) stand then for the fraction of the total number of modes (contained in the Brillouin zone of radius $Q_B$) corresponding to those in the normal and condensate contributions, respectively. Moreover, $\beta_c < \beta_n$ (in general much smaller) since the privileged modes to be largely excited are the minority ones at the lowest frequencies (see for example [39]).

We solve the stationary equations of evolution, to obtain the expression for the populations given in Appendix C. Numerical solutions are derived introducing values for the different parameters that appear in the basic Eqs. (19a and 19b). For illustrative purposes only, we choose those corresponding to the polar semiconductor GaAs, namely, $Q_B = 5.6 \times 10^7$ cm$^{-1}$; the velocity of sound in the thermal bath $s = 5 \times 10^5$ cm/s$^{-1}$; $\omega_n = 5.4 \times 10^{13}$ s$^{-1}$; $\omega_c = 4.5 \times 10^{13}$ s$^{-1}$; $\tau_n \simeq \tau_c \simeq 10^{-11}$ s; further we take $T_0 = 300$ K. The open parameters in the calculations are $\beta_n$, and $\beta_c$, and the quantity $\lambda$. We take a variable pumping intensity $S_\lambda$, but to have a better characterization of the phenomenon we put $S_\lambda = 0$, i.e., the modes lowest in frequency are not excited by the external source.

In Eqs. (C.1a and C.1b) we take $\beta_n = 0.9$ and $\beta_c = 0.1$, i.e., 90% of the modes higher in frequency transfer energy to 10% of the modes of lower frequencies. Moreover, we explore the influence of the parameter $\lambda$, which measures the strength of the nonlinear coupling term (for GaAs it can be grossly estimated as being of order one), taking for it the values $\lambda = 1.0$ in Fig. 1a and $\lambda = 10^{-5}$ in Fig. 1b. The apparent discontinuity in the slope in the curve for the higher mode population $\tilde{v}_n$ in Fig. 1b, near the point of crossover with the curve for $\tilde{v}_c$, is just an artifact resulting from a very rapid change in a very short interval of $S_\lambda$, not properly accounted for by the computational program.

Fröhlich effect is clearly evidenced: After a sufficiently intense pumping intensity is reached, which we call $S_\lambda^0$, there follows a very steep (near ‘explosive’) increase of the population of the modes lowest in frequency, while a saturation of the ‘pumping’ modes is observed. We noticed that it follows for practically any nonnull value of the nonlinear coupling strength $\lambda$, in other words, the complex behavior there occurs whenever the nonlinear contribution to the kinetic equations of evolution is present. In the figures we show the two cases where the two $\lambda$ differ by five orders of magnitude. It can be noticed that they differ in the values of the critical intensity for the onset of the phenomenon, the figures showing a separation of four orders of magnitude (say, roughly $S_\lambda^0 \sim 20$ for $\lambda = 1.0$ and $S_\lambda^0 = 4 \times 10^5$ for $\lambda = 10^{-5}$).

In Figs. 2a and b we have drawn the quasi-chemical potential as defined by Eq. (15), of the representative mode with the lowest frequency, in both cases (those of Figs. 1a and b, respectively). It follows that they tend asymptotically to the energy of the mode as $S_\lambda$ tends to infinity, but, otherwise, they always remain slightly below that value for $S_\lambda$ larger than the critical value.

Finally in Figs. 3a and b are shown the quasi-temperatures as defined by Eq. (17) (in units of $T_0$) in both cases, where is also clearly evidenced the steep increase of this quantity around the value $S_\lambda$. This case is usually referred to as production of ‘hot phonons’ [34–36].
Fig. 1. The steady-state populations of the pumped modes, $v_n$, and of the modes lowest in frequency, $v_e$, as a function of the (scaled) intensity of the source $S_n$ are shown using $x_n = 0.9$, $x_e = 0.1$, $\omega_m = 5.4 \times 10^{13}$ s$^{-1}$, $\omega_c = 4.5 \times 10^{13}$ s$^{-1}$, and $T_0 = 300$ K, in panel (a) for $\lambda = 1.0$ and in (b) for $\lambda = 10^{-5}$.

Fig. 2. The dependence on the intensity of the source of the quasi-chemical potential $\mu$, in units of $\hbar \omega_c$, corresponding to the populations $v_i$ in Fig. 1.

Fig. 3. The dependence on the intensity of the source of the quasi-temperature $T^*$ in units of $T_0$ corresponding to the populations $v_i$ in Fig. 1.
4. Summary and concluding remarks

We have analyzed the nonequilibrium macroscopic state of a system of excited (by the action of an external energy-pumping source) vibrational modes, that are in contact (through an anharmonic interaction) with a thermal bath of lower-lying-in-frequency vibrational acoustic-like modes. For that purpose we resorted to the use of the nonlinear quantum transport theory derived from the nonequilibrium statistical operator method. High order relaxation effect – up to fourth order in the interaction strengths – were introduced, which are those that ensure the emergence of complex behavior in the system.

We have explicitly obtained the equations of evolutions for the populations of the vibrational modes, being able to show that bilinear terms can produce the remarkable effect of transferring, in a cascade-like process, the energy that the different modes are receiving to the modes with the lowest frequencies. In a formal writing is introduced a Bose–Einstein-like distribution of the vibrational modes, characterized by the temperature of the bath and a quasi-chemical potential for each mode. The latter is zero in equilibrium (absence of the external source), as it should to produce the well-known Planck distribution, but becomes nonnull and increasing with increasing source power. Hence, the one for the mode of lowest frequency may approach this frequency leading to a ‘runaway’ in the population of such mode.

A numerical solution for a model system is described in Section 3. We were able to show that, in fact, there exists a critical value of the pumping power beyond which it is produced, and an enormous increase in the population of the modes lowest in frequency at the expenses of all the other modes. As shown, the quasi-chemical potentials of such modes tend asymptotically (as the source intensity goes to infinity) to coincide with the value of the modes’ frequencies. The populations of the modes lowest in frequency increase enormously, while those of all the other modes achieve almost saturation. Hence, there follows a kind of Bose–Einstein condensation in the sense that the distribution in the modes corresponds to a large accumulation in the states of lowest energies. But it should be emphasized that this occurs in a dissipative structure (nonequilibrium conditions) after a critical pumping intensity is achieved.

The dependence of the phenomenon on the properties of the system and its main characteristics have been discussed in Section 3. Here we only emphasize that this unexpected complex behavior arises as a result of the nonlinear characteristics of the kinetic equations in the nonequilibrium dissipative state of the system. As indicated in Section 1 we call the phenomenon Fröhlich effect [29–31], of which ours is a detailed calculation, in terms of a statistical nonequilibrium thermodynamics, invoking high order relaxation effect.

Concerning real systems where the situation we have described here may be present, we can mention two cases. One is that of polar semiconductors where there are high frequency optical modes and low frequency acoustic modes (see for example [40–42]). Another case is that of dipolar vibrations in biopolymers and other biological materials, excited by metabolic processes, and in contact with a thermal bath as those in [28–31]. Exact model calculations [39] seem to show that biopolymers are quite appropriate candidates for the actual occurrence of the Fröhlich effect, and the case of the molecular polymer acetalanilide has been considered in [43]. Moreover, the kind of nonlinearities we are considering here seems to lead to particular complex behavior (Fröhlich effect, Davydov soliton, Cherenkov-like radiation) in the propagation of ultrasound waves in biomaterials [44,45].

Finally, we stress that, as noticed before, the behavior of the system is even more complex, in the sense that beyond the critical intensity for the emergence of Fröhlich effect, excitations in the system at the lowest frequency propagate in a coherent fashion, with almost no dissipation, and being of the solitary wave type [43–45]. It is worth mentioning that the characteristics of the curve of population versus intensity of the pumping source, together with the few frequencies involved (near ‘monocromaticism’), coherent and dissipationless propagation of the excitations, are formally similar with the phenomenon of the laser effect. Another case is the so-called ‘exciton’, or spontaneous amplification of low-energy excitons populations in a coherent state [46,47]. Additional considerations on the Fröhlich effect are given in [48–50].

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Appendix A. Details of Eq. (12)

In Eq. (12) the several coefficients are given by

\[
\frac{1}{\tau_k} = \frac{4\pi}{\hbar^2} \sum_{q} |V_{kq}|^2 \xi^2 \delta(\Omega_{k-q} + \Omega_q - \omega_k) + \frac{4\pi}{\hbar^2} \sum_{q} |V_{kq}|^2 \xi^2 \delta(\Omega_{k+q} - \Omega_q - \omega_k),
\]

plays the role of the reciprocal of a relaxation time, and

\[
M^{(1)}_{kq} = \frac{4\pi}{\hbar^2} |V_{kq}|^2 \xi^2 \delta(\Omega_{k+q} - \omega_q - \omega_k),
\]

\[
M^{(2)}_{kq} = \frac{4\pi}{\hbar^2} |V_{kq}|^2 \xi^2 \delta(\Omega_{k+q} + A_{kq}),
\]

\[
A_{kq} = A^{(1)}_{kq} + A^{(2)}_{kq},
\]

with

\[
A^{(1)}_{kq} = \frac{8\pi}{\hbar^2} \sum_{q} |V_{kq}|^2 |V_{q'k}|^2 \xi^2 \delta(\Omega_{q'k} + \Omega_{q+k} - \omega_k - \omega_q) \left\{ |\Omega_{q'-q} + \Omega_{q+k} - \omega_k|^2 + |\Omega_{q'-k} - \Omega_q - \omega_k|^2 + |\Omega_{q-k} + \omega_q - \omega_k|^2 \right\} \delta(\Omega_{q'-k} - \Omega_q - A_{kq}),
\]

\[
A^{(2)}_{kq} = \frac{8\pi}{\hbar^2} \sum_{q} |V_{kq}|^2 |V_{q'k}|^2 \xi^2 \delta(\Omega_{q'-k} + \Omega_{q+k} - \omega_k - \omega_q) \left\{ |\Omega_{q'-q} + \Omega_{q+k} - \omega_k|^2 + |\Omega_{q'-k} - \Omega_q - \omega_k|^2 + |\Omega_{q-k} + \omega_q - \omega_k|^2 \right\} \delta(\Omega_{q'-k} + \Omega_q - A_{kq}).
\]

It should be noted that the terms with coefficients \( M \) are contributions from \( J^{(2)} \) arising from the anharmonic interactions \( H_{22} \) and \( H_{23} \), respectively; those with coefficients \( A \) come from \( J^0 \). Further,

\[
A_{kq} = \omega_k - \omega_q,
\]

\[
v_k^0 = [\text{e}^{\hbar \omega_k} - 1]^{-1},
\]

is the population in equilibrium of the pumped modes, and we have expressed the time-dependent correlations involving the operators associated to the source in terms of a spectral density, namely,

\[
\frac{2\pi}{\hbar} \langle \phi_k \phi_k^+(t) \rangle = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega I_k(\omega)e^{i\omega t},
\]

where \( I_k \) is the intensity of the source at frequency \( \omega \) in its Fourier spectrum.

Appendix B. The expression for the quasi-chemical potential

After some algebraic steps, we can obtain from Eq. (12), once the steady state is achieved and Eq. (16) is introduced, that

\[
\exp \{ \beta(\hbar \omega_k - \mu_k) \} = \frac{N_k}{D_k},
\]
where
\[ N_k = I_k + \frac{\psi_0}{\tau_k} + \bar{v}_k, \]  \hspace{1cm} (B.2a)
\[ D_k = I_k + \frac{\psi_0}{\tau_k} + \psi_k, \]  \hspace{1cm} (B.2b)
\[ \bar{v}_k = \frac{1}{\tau_k} + \sum_q M_{kq}^{(1)} v_q e^{\theta_{kq}} + \sum_q M_{kq}^{(2)} (1 + v_q) e^{\phi_{kq}} + \sum_q A_{kq} (1 + v_q) e^{i\phi_{kq}}, \]  \hspace{1cm} (B.2c)
\[ \psi_k = \sum_q \left[ M_{kq}^{(1)} (1 + v_q) + M_{kq}^{(2)} v_q + A_{kq} v_q \right]. \]  \hspace{1cm} (B.2d)

Appendix C. The stationary population

We rewrite Eqs. (19a) and (19b), using Eq. (20) and as a scaling factor the relaxation time \( \tau_c \), to obtain
\[ S_n - \left( \bar{v}_n - v_n^0 \right) - \frac{2}{\tau_n} \tilde{v}_n \left( 1 + \bar{v}_n \right) e^{i\phi_{n\infty}} - \bar{v}_n \left( 1 + \bar{v}_n \right) = 0, \]  \hspace{1cm} (C.1a)
\[ S_c - \frac{\tau_n}{\tau_c} \left( \bar{v}_c - v_c^0 \right) + \frac{2}{\tau_n} \tilde{v}_n \left( 1 + \bar{v}_n \right) e^{i\phi_{n\infty}} - \bar{v}_n \left( 1 + \bar{v}_n \right) = 0, \]  \hspace{1cm} (C.1b)
where the bar over the populations indicates a steady-state value, \( S_{n(c)} = \tau_n I_{n(c)} \), \( \tau_{n(c)} \) is defined in Eqs. (21a) and (21b), and \( \lambda = (\gamma Q_{\infty}^2 / 6 \pi^2) A_{\infty} \tau_n \). Using Eq. (C.1a) in Eq. (C.1b) it follows that
\[ a_1 \bar{v}_c^2 + a_2 \bar{v}_c + a_3 = 0, \]  \hspace{1cm} (C.2)
where
\[ a_1 = \frac{\lambda \tau_n}{\lambda \tau_c}, \]  \hspace{1cm} (C.3a)
\[ a_2 = \frac{1}{z} \left[ a_1 e^{i\phi_{n\infty}} + \frac{\tau_n}{\tau_c \lambda \lambda} + 1 \right] - A, \]  \hspace{1cm} (C.3b)
\[ a_3 = -\frac{1}{z} \left[ A e^{i\phi_{n\infty}} + \frac{1}{\lambda \lambda} \left( S_c + \frac{\tau_n}{\tau_c} v_c^0 \right) \right], \]  \hspace{1cm} (C.3c)
\[ z = e^{i\phi_{n\infty}} - 1 \]  \hspace{1cm} (C.3d)
and
\[ A = S_n + \frac{\tau_c}{\tau_n} S_c + v_n^0 + a_1 v_c^0. \]  \hspace{1cm} (C.3e)

If we take parameters characteristic of GaAs, it follows that \( v_n^0 \approx 0.34 \) and \( v_c^0 \approx 0.47 \), at 300 K. Next, since we are interested to evidence the expected complex behavior that follows after a certain threshold in the intensity of the pumping source has been attained, which implies that \( \tau_n I_n \) is large, we find that
\[ \bar{v}_c = \frac{1}{a_1} \left[ S_n + \frac{\tau_c}{\tau_n} S_c + v_n^0 + a_1 v_c^0 \right] - \frac{1}{z} \left( 1 + \frac{\tau_n}{\tau_c \lambda} \right), \]  \hspace{1cm} (C.4)
and that
\[
\bar{v}_n = s_n + \frac{\tau_e}{\tau_n} s_c + v_0^0 + v_1^0 - a_1 \bar{v}_e = \frac{1}{z} \left( 1 + \frac{\tau_n}{\tau_c s_n} \right),
\]  
(C.5)

Clearly, Eqs. (C.4) and (C.5) show that for a sufficiently intense pumping source (as noted, above a certain threshold, the populations of the modes in the condensate increase largely, even with \( s_c = 0 \), while the populations of the modes higher in frequency, however being constantly pumped, remain constant. This indicates that all the energy pumped on the system is transferred to the modes lowest in frequency, leading to the said nonequilibrium Bose–Einstein condensation. It may be noticed that the value of \( s_n z \) determines the threshold for the onset of the Fröhlich effect, but is not relevant beyond that point for determining the population in the condensate, since the first term on the right-hand side of Eq. (C.4) is much larger than the sum of the others.

References


