Complex Behavior in the Nonlinear Quantum Kinetics of Coupled Vibrational Modes

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Receivedl August 7, 1995

A model of coupled vibrational modes, interacting through anharmonic effects and subject to the action of an external source of energy that drives the system away from equilibrium, is considered. It is shown that the nonlinear equations that describe the dissipative evolution of the macroscopic state of the system admit a solution which evidences complex behavior in such system. It consists in the emergence of a phenomenon that resembles the Bose-Einstein condensation in the ideal gas of bosons in equilibrium at low temperatures, but in this case in nonequilibrium conditions, and, then, it implies in a transition between dissipative structures in Prigogine's sense. This paper presents a detailed extension of a short communication to appear in Physica D.

I. Introduction

Nonlinearity in physical, chemical, and biological systems is the source of new and unexpected complex behavior. Complexity manifests itself particularly in two lrind of situations related to dynamical systems: One is chaotic behavior in mechanical systems, where the idea that a system can be both deterministic yet unpredictable is a novelty with healthy development in recent years. 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]; 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^[2,3], synergetics^[4], and macroconcepts^[5].

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

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, mechano-statistical formalism, namely, the Nonequilibrium Statistical Operator Method (NSOM). It has been the object of several approaches which, as we have shown^[6], can be placed within the context of a unifying variational procedure based on Javnes' Predictive Statistical Mechanics^[7]. The NSOM allows for the construction of a nonlinear quantum transport theory - a far-reaching generalization of the Chapman-Enskog's and also Mori's methods - that describes the evolution of the system at the macroscopic level in arbitrar~nonequilibrium situations^[8]. Among the different NSOM we resort here to the use of Zubarev's approach^[9] and what we have called fourth order approximation in the theory of relaxation (FOART for short)^[8], which introduces the nonlinearities responsible for the complex behavior of the system of nonequilibrium oscillators.

We derive the equations of evolution for the population of the vibrational modes characterized by a frequency dispersion relation. We take a periodic distribution of the oscillating centers, and then the wave vector in the dispersion relation runs over a Brillouin zone, like, for example, polar modes in solids^[10] or dipolar vibration centers in biopolymers^[11]. 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.

In the next sections, after the derivation and discussion of the equations of evolution provided by the NSOM-FOART 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^[12] (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). Excitations are then accumulated in these modes largely increasing the value of their populations. In that way we have a steepily increasing population in the vibrational states with the lowest energy at the expenses of those with larger energies; there occurs then a phenomenon akin to a Bose-Einstein condensation, not for phases in equilibrium, but in nonequilibrium dissipative structures. It should be stressed 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.

The paper is organized as follows: in the next section it is presented a brief review of the theoretical background to be used in the calculations. In Section III we derive the equations of evolution for the vibrational mode populations, and their general aspects are discussed. In Section IV we proceed to the presentation of numerical calculations that clearly characterize the results. In the last section we briefly review and comment the results.

II. Theoretical background

The NSOM can be considered as a generalization of the statistical formalisms based on Boltzmann and Gibbs fundamental ideas. Different approaches have been developed by several authors, relying on either heuristic arguments, or using projection operator techniques. A unifying approach based on a variational principle is described in Ref. [6].

The NSOM is based, in any of its formulations, on Bogoliubov's assertion (principle of correlation weakening)^[13] that in general there exists a hierarchy of relaxation times such that as time goes on the system keeps loosing memory of the previous evolution, so that an ever decreasing number of variables is enough for the description of the macroscopic state of the system. This contraction is connected with the separation from the total Hamiltonian of strong interactions with certain symmetries^[14]; these interactions are those related to the fast relaxing processes. In the contracted description, the macroscopic state of the system is characterized by a reduced set of thermodynamic variables, say $Q_j(t)$ with j = 1, 2, ..., n, which are the average values with the nonequilibrium statistical operator (NSO) of a corresponding set of dynamical variables P_j , the NSO being a functional of these and only these variables. The clioice of these variables is not unique and one of the fundamental questions of the theory consists in defining in some sense their completeness^[15]. One way to perform such choice, associated to the NSOM, which is shown to be closely connected with phenomenological irreversible thermodynamics^[16], is based on the separation of the total Hamiltonian into two parts, namely

$$H = H_0 + H' , \qquad (1)$$

where H_0 contains the linetic energies and the part of the interactions that produce very rapid relaxation processes, and H' is related to the slow relaxation processes. Further, quantities P_j and the relevant part of the Hamiltonian, H_0 , are connected by what we term Peletmiiisliii-Ziibarev's symmetry condition, nainely

$$[P_j, H_0] = \sum_{k=1}^n \alpha_{jk} P_k , \qquad (2)$$

in an appropriate quaiitum representatioii, and where α_{jk} are c-numbers. In this way it may be said that the fast relaxing variables have been eliminated froin the clescription and the macrostate of the system is characterized in terms of the contracted description generated by the set of slow relaxing variables.

As already mentioned, the NSOM can be brought under a unifying approach resorting to a variational principle, namely Jaynes' principle of maximization of information entropy^[7], with memory effects and ad hoc hypothesis^[6]. The process consist in maximizing Gibbs functional

$$S_G(t) = -Tr\{\rho(t)\ln\rho(t)\},\qquad(3)$$

where ρ is the NSO, subject to the constraints that the set of macrovariables $Q_j(t)$ are those that properly describe the macroscopic state of the system, and that

$$Q_j(t') = Tr\{P_j\rho(t')\}$$
(4)

where $t_0 \leq t' \leq t$, with t_0 being the initial time of preparation of the system and t the time a measurement is performed. Eq. (4) introduces a dynamical character in the information (after-effects) since these coilditions involve the evolution of the system from the initial time of preparation t_0 (to be understood as much larger than the relaxation times in Bogoliubov's hierarchy associated to the principle of correlation weakening) up to time t. Tlie formal character of Eqs. (4) must be noticed, where one makes the assumption that there is a knowledge of the values of variables Q_i on tlie time interval (t_0, t) . However, this informationgathering interval can, and should, be reduced to inforination recorded at a unique time: the formalism produces equations of evolution for variables $Q_i(t)$ which give their values at any time $t > t_0$, once initial values $Q_i(t_0)$ provided.

We omit the details of the description of the variational procedure, and refer the reader to the work of Ref. [6]. It suffices to say that the Lagrange multipliers introduced by the variational method introduces are specified in a special way, in order: (1) to fix an initial condition from which proceeds the irreversible evolution of the macroscopic state of the nonequilibrium many-body system, what introduces from the outset a condition for dissipativity in an ad hoc manner; (ii) to introduce a set of functions $F_j(t)$ such that they have the role of intensive variables thermodynamically conjugated to the intensive variables $Q_j(t)$, to generate complete connection with phenomenological nonequilibriuin thermodynaniics; and (iii) to separate the NSO into two parts

$$\rho(t) = \bar{\rho}(t) + \rho'(t) , \qquad (5)$$

where the first term, $\bar{\rho}(t)$, is an auxiliary generalized Gibbsian distribution which defines the instantaneous values of the macrovariables, and $\rho'(t)$ carries the information on the microscopic dynamics relevant to the description of the irreversible evolution of the macroscopic state of the system. The auxiliary distribution is given by

$$\bar{\rho}(t) = \exp\{-\phi(t) - \sum_{j=1}^{n} F_j(t) P_j\}, \qquad (6)$$

where

$$\phi(t) = \ln Tr \left\{ \exp\left(-\sum_{j=1}^{n} F_j(t) P_j\right) \right\} , \quad (7)$$

ensures its normaliúation.

Conditiori (ii) above, placed on the variational Lagrange multipliers, requires the additional property

$$Q_j(t) = Tr\{P_j\rho(t)\} = Tr\{P_j\bar{\rho}(t)\} , \qquad (8)$$

which also provides for the simultaneous normalization of both $\rho(t)$ and $\bar{\rho}(t)$, (namely Eq. (7) in the last case), and makes of $\phi(t)$ a generating functional in the sense that

$$\frac{\delta\phi(t)}{\delta F_j(t)} = -Q_j(t) , \qquad (9)$$

defines the conjugation of both kind of variables (F and Q) in the context of phenomenological irreversible thermodynamics^[6,16].

Particular cases of the NSOM given in the literature are recovered within the variational method [Cf. Ref. 6], in particular Zubarev's NSO^[9], which is to be used in our analysis of the system of nonequilibrium oscillators in next section. It is given by

$$\rho_{\epsilon}(t) = \exp\left\{\epsilon \int_{-\infty}^{t} dt' e^{\epsilon(t'-t)} \ln \bar{\rho}(t',t'-t)\right\} , \qquad (10)$$

where $\bar{\rho}$ is given by Eq. (6) with the first time in the argument referring to the time dependence of the thermodynamic variables $F_j(t')$ and the second stands for the time evolution of operators $P_j(t'-t)$ in Heisenberg's representation. ϵ is a positive infinitesimal that

ensures the irreversible evolution of the system from initial preparation, and goes to zero after the trace operation in the calculation of average values has been performed. Integration by parts in Eq. (10) allows to rewrite Zubarev's NSO in the form

$$\rho_{\epsilon}(t) = \exp\left\{\ln\bar{\rho}(t,0) - \int_{-\infty}^{t} dt' e^{\epsilon(t'-t)} \frac{d}{dt'} \ln\bar{\rho}(t',t'-t)\right\} , \qquad (11)$$

which can be put in the form of Eq. (5) [6,9]. It is worth noticing that Zubarev's NSO satisfies a modified Liouville equation of the type

$$\left[\frac{\partial}{\partial t} + i\mathcal{L}\right] \ln \rho_{\epsilon}(t) = -\epsilon \left[\ln \rho_{\epsilon}(t) - \ln \bar{\rho}(t, O)\right], \qquad (12)$$

i.e. a Liouville equation (L is the Liouvillian of the system) with an infinitesimal source that breaks the, otherwise valid, time reversal symmetry. We note that the initial condition for the NSO is

i.e. the state characterized by the initial values $Q_j(-\infty)$ of the macrovariables with no correlation among them; for $t > t_0(=-\infty)$ the term $\rho'(t)$ is present and with it the irreversible evolution and correlation of the variables under the dynamics generated by the system Hamiltonian.

$$\rho_{\epsilon}(-\infty) \equiv \bar{\rho}(-\infty, 0) , \qquad (13)$$

The NSOM seems to be a powerful mechanostatistical formalism for the treatment of systems arbitrarily away from equilibrium. In particular it provides mechano-statistical foundations for phenomenological irreversible thermodynamics^[16]. Also, within its scope^[6], Glansdorff-Prigogine thermodynamic criterion of evolution, Prigogine's theorem of minimum entropy production, and Glansdorff-Prigogine (in)stability criterion are verified^[2]. Further, within the framework of the NSOM it is possible to construct a response function theory for far-from-equilibrium systems and an accompanying nonequilibrium thermodynamic Green function formalism^[6]. But the most important part in the NSOM is the construction of a nonlinear quantum transport theory for the basic variables, fundamental in all applications since they give the description of the irreversible evolution of the macroscopic state of the system. We have

$$\frac{d}{dt}Q_j(t) = Tr\left\{\frac{1}{i\hbar}\left[P_j, H\right]\rho_\epsilon\right\} ,\qquad(14)$$

and using the separation of Eq. (5) after some mathematical manipulations we are left with the equation^[17]</sup>

 $\frac{d}{dt}Q_j(t) = J_j^{(0)}(t) + J_j^{(1)}(t) + J_j^{(\text{coll})}(t) , \qquad (15)$

where

$$J_{j}^{(0)}(t) = Tr\left\{\frac{1}{i\hbar}[P_{j}, H_{0}]\bar{\rho}(t, 0)\right\} , \qquad (16a)$$

$$J_{j}^{(1)}(t) = Tr\left\{\frac{1}{i\hbar}[P_{j}, H']\bar{\rho}(t, 0)\right\} , \qquad (16b)$$

and

$$J_{j}^{(\text{coll})} = \left(\frac{1}{i\hbar}\right) \int_{-\infty}^{t} dt' e^{\epsilon(t'-t)} \left[Tr\left\{\frac{1}{i\hbar}[H, [H'(t-t'), P_{j}(t-t')]]\bar{\rho}(t', 0)\right\} + \sum_{k} \frac{dF_{k}(t')}{dt'} \frac{\delta Tr\{[H'(t-t'), P_{j}(t-t')\bar{\rho}(t', 0))}{\delta F_{k}(t')} \right]$$
(16c)

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and organized in increasing powers n of the interaction strengths, namely

$$\frac{d}{dt}Q_{j}(t) = \sum_{n=0}^{\infty} J_{j}^{(n)}(t) , \qquad (17)$$

with the construction of quantities $J_j^{(n)}$ described in Ref. [8].

The form of the collision operator given by Eq. (17) permits to introduce approximations by means of a truncation of the series of partial collision operators in a given order of the interaction. The lowest order that introduces relaxation effects is a truncation in second order in the interaction strengths: it renders the equations Markovian in character and we have called it^[8] the second order approximation in relaxation theory, SOART for short. It is usually referred to in the liter-

Eq. (15) is a far-reaching generalization of Mori's equations^[18]. The first two terms, $J^{(0)}$ and $J^{(1)}$ are, in Mori's terminology, precession terms, and the collision operator of Eq. (16c) is, differently to Mori's theory, a highly nonlinear term in the basic variables; also it is nonlocal in space (for simplicity we have omitted the possible space dependence of quantities P), and it contains memory effects.

The collision operator of Eq. (16c) is extremely difficult to handle in practical calculations. In Ref. [8] we show that, using the properties defined by Eqs. (1) and (2), the complicated collision operator of Eq. (16c) can be rewritten through the use of an appropriate operator for the propagation of the past history of the motion in terms of an infinite series of collision integrals which are instantaneous in time (given as averages over the auxiliary NSO of Eq. (6) at the time of measurement) ature as the linear theory of relaxation^[19], a name we avoid because of the misleading term linear that indicates a certain approximation in operator p in Eq. (5) and in the expression for the information-entropy production in the NSOM. In SOART the equations of evolution are a set of coupled and, in principle, highly nonlinear integrodifferential equations $[J^{(0)} \text{ and } and J^{(1)}$ are given by Eqs. (16), while $J^{(2)}$ is in Ref. [8] given by Eq. (62a)].

As remarked in the Introduction, for the treatment of the problem of studying nonlinear dissipative harmonic oscillators, we resort to the fourth order approximation FOART, which introduces, besides $J^{(0)}$, $J^{(1)}$, and $J^{(2)}$, the partial collision operators $J^{(3)}$ and $J^{(4)}$, given by Eqs. (62b), (62c) and (66) in Ref. [8].

We call the attention to the fact that the different $J^{(n)}$, with $n \ge 2$, are composed of several types of terms that can be summarized as being associated to three kind of contributions, namely: (a) the equivalent of the rionequilibrium statistically averaged contribution of the Born series in perturbation theory; (b) terms that carry the effect of the change in the nonequilibrium macrostate of the system during the irreversible evolution; and (c) terms that involve the effect of the past history of evolution (memory effects).

The partial collision operators $J^{(n)}$ are more and more intrincate with increasing n, producing a large number of contributions of the three types just mentioned. We anticipate that in the calculation up to n = 4 for the equations of evolution of the populations of the vibrational modes in the system being considered, because of the characteristics of the Hamiltonian and of the basic macrovariables, only contributions of the type (a) are present.

III. 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 $\omega_{\vec{q}}$, and a low frequency branch (acousticlike branch) with frequency dispersion relation $\Omega_{\vec{q}}$. The wave-vector \vec{q} runs over a 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 through a good thermal contact with a reservoir.

We write for the system Hamiltonian

$$H = H_0 + H'$$
, (18)

according to the requirement of the NSOM [Cf. Eq. (1)], where

$$H_0 = \sum_{\vec{q}} \hbar \omega_{\vec{q}} \left(a_{\vec{q}}^{\dagger} a_{\vec{q}} + \frac{1}{2} \right) + \sum_{\vec{q}} \hbar \Omega_{\vec{q}} \left(b_{\vec{q}}^{\dagger} b_{\vec{q}} + \frac{1}{2} \right)$$
(19)

and

$$H' = H'_f + H'_1 + H'_2 , \qquad (20)$$

with

$$H'_f = \sum_{\vec{q}} \varphi_{\vec{q}} a^{\dagger}_{\vec{q}} + H.C. , \qquad (21)$$

$$H_1' = \sum_{\vec{q}\vec{q}'} V_{\vec{q}\vec{q}'} a_{\vec{q}} b_{\vec{q}'}^{\dagger} b_{\vec{q}-\vec{q}'}^{\dagger} + H.C. , \qquad (22)$$

$$H_2' = \sum_{\vec{q}\vec{q}'} V_{\vec{q}\vec{q}'} a_{\vec{q}} b_{\vec{q}'} b_{\vec{q}+\vec{q}'}^{\dagger} + H.C. , \qquad (23)$$

In these equations $a(a^{\dagger})$ and $b(b^{\dagger})$ are the annihilation (creation) operators of the 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'_{f} accounts for the interaction between the pumping source and the vibrational modes, with φ and φ^+ being annihilation and creation operators for the excitations in the source, also incorporating the coupling strength. The other two terms, H'_1 and H'_2 are the contributions of the anharmonic interactions between both types of vibrations that will contribute to the equations of evolution: they correspond to the decay of one vibrational mode into two of the thermal bath, and of one vibrational mode and one of the thermal bath into one of the latter (and their Hermitian conjugates). Momentum conservation has been taken into account.

To deal with this system in NSOM the first step, as noted in Section II is to define the basic set of variables deemed appropriate for the description of its macroscopic state. We choose the population of the vibrational modes,

$$\nu_{\vec{q}}(t) = Tr\{a^{\dagger}_{\vec{q}} \ a_{\vec{q}}\rho_{\epsilon}(t)\} , \qquad (24)$$

and the ltinetic energy of tlie thermal bath,

$$E_B(t) = Tr\left\{\sum_{\vec{q}} \hbar\Omega_{\vec{q}} \left(b^{\dagger}_{\vec{q}} \ b_{\vec{q}} + \frac{1}{2}\right)\rho_{\epsilon}(t)\right\} , \quad (25)$$

where $\rho_{\epsilon}(t)$ is Zubarev's NSO for this case, with the auxiliary NSOM-operator [Cf. Eq. (6)] given by

$$\bar{\rho}(t,0) = \exp\left\{-\phi(t) - \sum_{\vec{q}} F_{\vec{q}}(t)a^{\dagger}_{\vec{q}} \ a_{\vec{q}} - \beta H_B\right\} ,$$
(26)

where $\beta = 1/kT$, with T being the constant temperature of the thermal bath, aiid $F_{\bar{q}}(t)$ is the nonequilibrium thermodynamic parameter conjugated to the dynamical variable population of the vibrational inodes; H_B is the Hamiltonian of the free thermal bath; and $\phi(t)$ [Cf. Eq. (7)] is the Massieu-Planck functional ensuring the normalization of the NSO. It should be noted that the symmetry condition of Eq. (2) is satisfied, with all α being null.

We are left with the task of evaluating the equation of evolution for the population of the vibrational inodes, what we do resorting to the NSOM-FOART, that is to say an equation of evolution containing the partial collision integrals of Eq. (17) up to the fourth order only, namely

$$\frac{d}{dt}\nu_{\vec{q}}(t) = J_{\vec{q}}^{(0)}(t) + J_{\vec{q}}^{(1)}(t) + J_{\vec{q}}^{(2)}(t) + J_{\vec{q}}^{(3)}(t) + J_{\vec{q}}^{(4)}(t) ,$$
(27)

with the collision integrals $J^{(0)}$ and $J^{(1)}$ given by Eqs. (16a) and (16b), and tlie others, as noted in last section, given in Ref. [8] by Eqs. (62a), (62b), (62c) and (66), where, of course, quantity P_j is here $a_{\bar{q}}^{\dagger}a_{\bar{q}}$. We recall that tlie other basic variable, namely, the energy of tlie tliermal bath of Eq. (25) is assumed to be constant in time and determined by the temperature T, that of equilibrium with the reservoir.

The calculation of Eq. (27) is lengthy and laborious^[20]; without going into the details we note that because of the particular'forms of the Hamiltonian, the auxiliary NSO, and tlie basic variables, several contributions vanish, iiamely $J^{(0)}$, $J^{(1)}$, and $J^{(3)}$. Moreover, of the multiple possible contributions to $J^{(2)}$ and $J^{(4)}$, involving the three types (a), (b), and (c) described at tlie end of Section II, they simply reduce to those of type (a), namely, tlie equivalent of tlie nonequilibrium statistical average of the second and fourth order contributions to the Born series in quantum perturbation theory. We finally obtain,

$$\frac{d}{dt}\nu_{\vec{q}}(t) = I_{\vec{q}}(\omega_{\vec{q}}) - \frac{1}{\tau_{\vec{q}}} [\nu_{\vec{q}}(t) - \nu_{\vec{q}}^{0}] + \sum_{\vec{q}} \Lambda_{\vec{q}\vec{q}'} \{\nu_{\vec{q}'}(t)[1 + \nu_{\vec{q}}(t)] - \nu_{\vec{q}'}(t)[1 + \nu_{\vec{q}'}(t)]e^{\beta\hbar\Delta_{\vec{q}\vec{q}'}}\},$$
(28)

where

$$\tau_{\vec{q}}^{-1} = \frac{4\pi}{\hbar^2} \frac{1}{\nu_{\vec{q}}^0} \sum_{\vec{p}} |V_{\vec{q}\vec{p}}|^2 \nu_{\vec{p}}^B \nu_{\vec{q}-\vec{p}}^B \delta(\Omega_{\vec{q}-\vec{p}} + \Omega_{\vec{p}} - \omega_{\vec{q}}) + \frac{4\pi}{\hbar^2} \frac{1}{\nu_{\vec{q}}^0} \sum_{\vec{p}} |V_{\vec{q}\vec{p}}|^2 \nu_{\vec{p}}^B \nu_{\vec{q}+\vec{p}}^B e^{\beta\hbar\Omega_{\vec{p}}} \delta(\Omega_{\vec{q}+\vec{p}} + \Omega_{\vec{p}} - \omega_{\vec{q}}) , \qquad (29)$$

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

$$\Lambda_{\vec{q}\vec{q}} = \Lambda^{(1)}_{\vec{q}\vec{q}'} + \Lambda^{(2)}_{\vec{q}\vec{q}'} , \qquad (30)$$

with

$$\Lambda^{(1)}_{\vec{q}\vec{q}'} = \frac{8\pi}{\hbar^4} \sum_{\vec{p}} |V_{\vec{p}\vec{q}'}|^2 \nu^B_{\vec{p}} \nu^B_{\vec{p}+\vec{q}-\vec{q}'} e^{\beta\hbar\Omega_{\vec{p}}}$$

$$[|\Omega_{\vec{p}-\vec{q'}} + \Omega_{\vec{p}} - \omega_{\vec{q'}}|^2 + |\Omega_{\vec{p}+\vec{q}} - \Omega_{\vec{p}} - \omega_{\vec{q'}}|^2 + |\Omega_{\vec{p}} - \omega_{\vec{q'}} - \omega_{\vec{p}-\vec{q'}}|^2 + |\Omega_{\vec{p}-\vec{q'}} - \omega_{\vec{p}+\vec{q'}}|^2 + |\Omega_{\vec{q}-\vec{q'}} - \omega_{\vec{q}}|^2]\delta(\Omega_{\vec{p}+\vec{q}-\vec{q'}} - \Omega_{\vec{p}} - \Delta_{\vec{q}\vec{q'}}) ,$$

$$(31)$$

$$\begin{split} \Lambda_{\vec{q}\vec{q}'}^{(2)} &= \frac{8\pi}{\hbar^4} \sum_{\vec{p}'} |V_{\vec{p}\vec{q}'}|^2 |V_{\vec{p}\vec{q}'}|^2 \nu_{\vec{p}}^B \nu_{\vec{p}+\vec{q}-\vec{q}'}^B e^{\beta\hbar\Delta_{\vec{q}\vec{q}'}} \\ [|\Omega_{\vec{p}-\vec{q}'} + \omega_{\vec{p}} - \omega_{\vec{p}+\vec{q}-\vec{q}'}|^2 + |\Omega_{\vec{p}} + \omega_{\vec{p}-\vec{q}'} - \omega_{\vec{q}'}|^2 + \\ + |\Omega_{\vec{q}-\vec{q}'} + \omega_{\vec{q}} - \omega_{\vec{q}'}|^2] \delta(\Omega_{\vec{p}+\vec{q}-\vec{q}'} + \Omega_{\vec{p}} + \Delta_{\vec{q}\vec{q}'}) \;. \end{split}$$
(32)

It should be noticed that the terms that appear in the expression for τ^{-1} [Eq. (29)] are contributions from $J^{(2)}$, while those with coefficieitis A come from $J^{(4)}$. Furthermore,

$$\Delta_{\vec{q}\vec{q}'} = \omega_{\vec{q}} - \omega_{\vec{q}'} , \qquad (33)$$

$$\nu_{\vec{p}}^{B} = [\exp(\beta \hbar \Omega_{\vec{p}}) - 1]^{-1} , \qquad (34a)$$

is the population in equilibrium of the thermal bath modes,

$$\nu_{\vec{q}}^{0} = [\exp(\beta \hbar \omega_{\vec{q}}) - 1]^{-1} , \qquad (34b)$$

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

$$\frac{2\pi}{\hbar} < \varphi_{\vec{q}} \varphi_{\vec{q}}^{\dagger}(t) > = \int_{-\infty}^{\infty} \frac{d\omega}{\pi} I_{\vec{q}}(\omega) e^{i\omega t} , \qquad (35)$$

where $I_{\vec{q}}$ is the intensity of the source at frequency w in its Fourier spectrum.

The equation of evolution, Eq. (28), for the population of the \vec{q} -mode, is composed of several contributions: the first is the one associated to the pumping effects (from the external source) that brings the system further and further away from equilibrium with increasing values of I (the intensity of the source); the second contribution accounts for the relaxation of the excess population, created by the source, to the thermal bath diminishing the value of the population; the one containing A (and arising from $J^{(4)}$) contains a nonlinear contribution, expressed by the product $\nu_{\vec{q}}\nu_{\vec{q}'}$, which can produce either a relaxation or an excitation effect depending on the sign of the difference $\omega_{\vec{q}} - \omega_{\vec{q}'}$. For a given mode \vec{q} this term tends to increase its population at the expense of the other modes \vec{q}' if it is verified that $\omega_{\vec{q}'} > \omega_{\vec{q}}$. 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 frequency in a cascading-like process.

With an external source acting continuously, after a transient time has elapsed a steady state must follow, i.e. $d\nu/dt = 0$. It should be stressed that the steady state in the absence of the source [I= 0 in Eq. (28)] is the equilibrium state, as can be verified using Eq. (28). Let us next look for the characteristics of this steady state. First, it is worth noticing that according to the NSOM [Cf. Eqs. (8)] in the present case it follows that

$$\nu_{\vec{q}}(t) = Tr\{a^{\dagger}_{\vec{q}}a_{\vec{q}}\bar{\rho}(t,0)\} = [e^{F_{\vec{q}}(t)} - 1]^{-1} .$$
 (36)

In equilibrium $F_{\vec{q}}^0 = \beta \hbar \omega_{\vec{q}}$ and Eq. (36) is the Planck distribution of the populations. Consider now the stationary nonequilibrium situation when, after some simple algebraic steps, we can obtain from Eq. (28) in the stationary state that the nonequilibrium intensive variable $F_{\vec{q}}$ can be written as

$$F_{\vec{q}} = \beta [\hbar \omega_{\vec{q}} - \mu_{\vec{q}}] , \qquad (37)$$

and, hence,

$$\nu_{\vec{q}} = \left[\exp\{\beta [\hbar \omega_{\vec{q}} - \mu_{\vec{q}}\} - 1\right]^{-1} , \qquad (38)$$

where the quantity $\mu_{\vec{q}}$ is defined by the expression

$$\exp\{\beta[\hbar\omega_{\vec{q}} - \mu_{\vec{q}}]\} = N_{\vec{q}}/D_{\vec{q}} , \qquad (39)$$

where

$$N_{\vec{q}} = I_{\vec{q}} + \nu_{\vec{q}}^0 \tau_{\vec{q}}^{-1} + \phi_{\vec{q}} , \qquad (40a)$$

$$D_{\vec{q}} = I_{\vec{q}} + \nu_{\vec{q}}^0 \tau_{\vec{q}}^{-1} + \psi_{\vec{q}} , \qquad (40b)$$

$$\phi_{\vec{q}} = \tau_{\vec{q}}^{-1} + \sum_{\vec{q}'} \Lambda_{\vec{q}\vec{q}'} (1 + \nu_{\vec{q}'} e^{\beta\hbar\Delta_{\vec{q}\vec{q}'}}) , \qquad (40c)$$

and

$$\psi_{\vec{q}} = \sum_{\vec{q}'} \Lambda_{\vec{q}\vec{q}'} \nu_{\vec{q}} \tag{40d}$$

Ea. (38) is an interesting alternative form of Eq. (28) in the stationary state: it is formally a Bose-Elnstein distribution with temperature β^{-1} and a quasi-chemical potential $\mu_{\vec{a}}$ for each mode. We call the attention to the fact that this quasi-chemical potential is a complicated functional of the population of all the modes. Further, the quasi-chemical potential per mode vanishes in the limit of vanishing pumping source, and then Eq. (38) becomes the Planck distribution in equilibrium, but with a non vanishing pumping source $\mu_{\vec{q}}$ is positive and growing with the increasing intensity of the source. Thus, 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 analize this possibility in next section on the basis of a simplified model. As a final word in this section we note that the concept of Bose-Einstein distributions with non-zero quasi-chemical potential in the nonequllibrium populations of bosons, that are otherwise Planckian in equilibrium, have been used by Landsberg for the populations of photons in the case of the steady state between radiation and an electronhole plasma in semiconductors^[21], and by Fröhlich for the characterization of the populations of dipolar wave excitations in biophysical systems^[12].

IV. Numerical solutions for a model system

The equations of evolution for the 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: Noticing the already referred effect that the modes at the lowest frequency receive the energy pumped on all the other higher frequency modes through the term

$$\Lambda_{\vec{q}\vec{q}'}(1-e^{\beta\hbar\Delta_{\vec{q}\vec{q}}})\nu_{\vec{q}'}\nu_{\vec{q}}, \qquad (41)$$

in a cascading-down process, we introduce a crude model consisting in concentrating the effect of the pumping modes in only one (say q = 0 at the highest frequency), associated with a degeneracy factor, g_0 , accounting in average for **all** the others, and the modes with the lowest frequency, (at the boundary zone) which we characterized by wave-vector \vec{Q} , and a degeneracy g_Q .

We verify the following properties:

$$\Lambda_{\vec{a}'\vec{a}} = \Lambda_{\vec{a}\vec{a}'} e^{\beta\hbar\Delta_{\vec{q}\vec{q}'}} , \qquad (42a)$$

and for \vec{Q} being a boundary zone vector there follows tlie "nesting" condition

$$\Omega_{\vec{p}\pm\vec{Q}} = \Omega_{\vec{Q}} - \Omega_{\vec{p}} , \qquad (42b)$$

and we introduce next a Debye model, namely we take $\Omega_{\vec{p}} = s |\vec{p}|$, where s is the group velocity of propagation. Finally we take the matrix elements $V_{\vec{q}\vec{p}}$ as depending only on the momentum transfer, and we write V(q) for them. After performing the integration in \vec{p} in the expression for $\tau_{\vec{q}}$ of Eq. (29) we find an expression for the matrix element in term of the relaxation time, the latter to be taken as a phenomenological parameter, namely

$$|V(\vec{q})|^2 = 2\pi\hbar^4 s^3 \nu_{\vec{q}}^0 \beta^2 / V \tau_{\vec{q}} , \qquad (43)$$

where V is the volume of the system. In the proposed model introducing Eq. (43) in the expression for the quantity A of Eq. (31) and, after that, performing the integrations in \vec{p} we find five vanishing contributions, namely those associated with $\Lambda^{(1)}$ given by

$$\Lambda_{0\vec{Q}}^{(1)} = \sum_{j=1}^{5} \lambda_{0\vec{Q}}^{(1)} .$$
(44)

In Eq. (44) we have introduced the quantities,

$$\lambda_{0\vec{Q}}^{(1)} = \frac{4\pi}{V} \frac{\nu_0^0 \nu_{\vec{Q}}^0}{\tau_0 \tau_{\vec{Q}}} h^4 s^3 \beta^4 f_{0\vec{Q}} \left| \frac{\Omega_{\vec{Q}} - \Delta_{0\vec{Q}}}{\Omega_{\vec{Q}} - \omega_{\vec{Q}}} \right|^2 , \qquad (44a)$$

where

$$f_{0\vec{Q}} = \nu_{0\vec{Q}}^{(+)} \nu_{0\vec{Q}}^{(-)} e^{\beta\hbar(\Omega_Q - \Delta_{0Q})/2} ,$$

with

$$\nu_{0\vec{Q}}^{(\pm)} = \left[\exp\left\{ \frac{1}{2}\beta\hbar(\Omega_{\vec{Q}} \pm \Delta_{0\vec{Q}}) \right\} - 1 \right]^{-1}$$

moreover,

$$\lambda_{0\vec{Q}}^{(2)} = (|\Omega_{\vec{Q}} - \omega_{\vec{Q}}|^2 / |\omega_0|^2) \left| \frac{\Omega_{\vec{Q}} - \omega_{\vec{Q}}}{\Omega_{\vec{Q}} - \Delta_{0\vec{Q}}} \right| \lambda_{0\vec{Q}}^{(1)} ,$$
(44b)

$$\lambda_{0\vec{Q}}^{(3)} = \left| \frac{\Omega_{\vec{Q}} - \omega_{\vec{Q}}}{\omega_{\vec{Q}} - \Delta_{0\vec{Q}}} \right|^2 \left| \frac{\Omega_{\vec{Q}} - \Delta_{\vec{Q}}}{\Gamma_{0\vec{Q}}^{(3)}} \right|^2 \lambda_{0\vec{Q}}^{(1)} , \qquad (44c)$$

where

$$\Gamma_{0\vec{Q}}^{(3)} = \omega_0 - (\Delta_{0\vec{Q}}/2s^2Q^2)(\Omega_{\vec{Q}} + \Delta_{0\vec{Q}})^2 - \frac{1}{2}(\omega_{\vec{Q}} - \omega_{\vec{0}} - \omega_{\vec{Q}}) , \qquad (44d)$$

also,

$$\lambda_{0\vec{Q}}^{(4)} = \left| \frac{\omega_{\vec{Q}} - \Delta_{0\vec{Q}}}{\Gamma_{0\vec{Q}}^{(4)}} \right|^2 \left| \frac{\Omega_{\vec{Q}} - \omega_{\vec{Q}}}{\Omega_{\vec{Q}} - \Delta_{0\vec{Q}}} \right|^2 \lambda_{0\vec{Q}}^{(1)} , \qquad (44e)$$

where

$$\Gamma_{0\vec{Q}}^{(4)} = \omega_0 - (\Delta_{0\vec{Q}}/2s^2Q^2)(\Omega_{\vec{Q}} - \Delta_{0\vec{Q}})^2 - \frac{1}{2}(\omega_{\vec{Q}} + \omega_{\vec{0}} + \omega_{\vec{Q}}), \qquad (44f)$$

and finally

$$\lambda_{0\vec{Q}}^{(5)} = \left| \frac{\Omega_{\vec{Q}} - \omega_{\vec{Q}}}{\Omega_{\vec{Q}} - \Delta_{0\vec{Q}}} \right|^2 \left| \frac{\Omega_{\vec{Q}} + \Delta_{0\vec{Q}}}{\Omega_{\vec{Q}} - \Delta_{0\vec{Q}}} \right|^2 \lambda_{0\vec{Q}}^{(1)} .$$

$$(44g)$$

The two coupled equations of evolution are then

$$\frac{d}{dt}\nu_0 = I_0 - \frac{1}{\tau_0}(\nu_0 - \nu_0^0) - g_{\vec{Q}}R_{0\vec{Q}} , \qquad (45a)$$

$$\frac{d}{dt}\nu_{\vec{Q}} = I_{\vec{Q}} - \frac{1}{\tau_{\vec{Q}}}(\nu_{\vec{Q}} - \nu_{\vec{Q}}^0) + g_0 R_{0\vec{Q}} , \qquad (45b)$$

where

$$R_{0\vec{Q}} = \Lambda_{0\vec{Q}} \{ \nu_0 (1 + \nu_{\vec{Q}}) e^{\beta \hbar \Delta_{0\vec{Q}}} - (1 + \nu_0) \nu_{\vec{Q}} \} .$$
(46)

It is worth noticing that Eqs. (45) are of the type of Lotka-Voltera's predator-prey equations^[22]. The degeneracy coefficients g are proportional to the extension of the system; we take for them the expressions

$$g_0 = \alpha_0 \frac{V}{(2\pi)^3} \frac{3}{4} \pi Q^3 , \qquad (47a)$$

$$g_Q = \alpha_Q \frac{V}{(2\pi)^3} \frac{3}{4} \pi Q^3 , \qquad (47b)$$

where α_0 and α_Q (with both smaller than one, and $\alpha_0 + \alpha_Q \leq 1$) stand then for the fraction of the num-

ber of the two types of modes that in our model are contained in the Brillouin zone.

To obtain a computational solution we need to introduce numerical values for the different parameters. For illustrative purposes only, we choose those corresponding to the polar semiconductor GaAs, namely, Q = $5.6 \times 10^7 \text{ cm}^{-1}$; $s = 5 \times 10^5 \text{ cm sec}^{-1}$; $\Omega_{\vec{Q}} = 4.3 \times 10^{13}$ sec^{-1} ; $\omega_0 = 5.4 \times 10^{13} \text{ sec}^{-1}$; $\omega_{\vec{Q}} = 4.5 \times 10^{13} \text{ sec}^{-1}$; $\tau_0 \simeq \tau_Q \simeq 10^{11} \text{ sec}$; further we take T = 300K. There are two open parameters in the calculations, namely, α_0 and α_Q . We solve the equations for different values of them, i.e. we test the dependence of the behavior of the system on them.

Using the values stated above for the different parameters we find that the two coupled equations (45) in the stationary state can be transformed in the two following equations, namely

$$\nu_Q^{-2} + [F(\alpha_0, \alpha_Q) - (\alpha_0/\alpha_Q)\tau_Q I_0]\bar{\nu}_Q -$$

$$-[G(\alpha_0, \alpha_Q) + 4.9(\alpha_0/\alpha_Q)\tau_Q I_0] = 0 , \qquad (48a)$$

$$\bar{\nu}_0 = \bar{\nu}_0^0 + \tau_0 I_0 - (\tau_0 \alpha_Q / \tau_Q \alpha_0) (\bar{\nu}_Q - \bar{\nu}_Q^0)$$
(48b)

where

$$F(\alpha_0, \alpha_Q) = 4.4 + 120\alpha_Q^{-1} + 3.6(\alpha_0/\alpha_Q) ,$$

$$G(\alpha_0, \alpha_Q) = 2.3 + 55.8\alpha_Q^{-1} + 1.6(\alpha_0/\alpha_Q)$$

and the bar in $\bar{\nu}$ indicates the stationary value of the population.

We have taken a variable pumping intensity I_0 , but for the purpose in what follows of a better characterization of the phenomenon we put $I_Q = 0$, i.e. the lowest frequency modes are not excited by the external source.

Fig. 1 shows the dependence of the steady-state populations of both types of modes with the pumping intensity, for the choice $\alpha_0 = 0.2$, $\alpha_Q = 10^{-4}$, while in Fig. 2 we display the case $\alpha_0 = 0.6$ and $\alpha_Q = 10^{-4}$.



Figure 1. The populations of the pumped modes, ν_0 , and of the lowest frequency modes, ν_Q , as a function of the intensity of the source. Parameters α (see text) are indicated in the upper left corner.

Fröhlich effect is clearly evidenced: after a sufficiently intense pumping intensity is reached, which we call I_0^c , there follows a very steep (near "explosive") increase of the population of the lowest frequency modes, while it is observed a saturation of the "pumping" modes.

Closing this section we look for some asymptotic forms, meaning $I_0 \gg I_0^c$, for the equations in the stationary state, that would help to clarify some of the numerical results. After the critical point has been surpassed, $\nu_{\vec{Q}}$ is very large; then $\nu_{\vec{Q}}$ is much larger than $\nu_{\vec{8}}^0$ and 1, and this is also valid for ν_0 . Hence, using Eqs. (45) we obtain a couple of equations for the stationary asymptotic populations $\bar{\nu}^A$, namely

$$I_0 - \tau_0^{-1} \bar{\nu}_0^A - g_Q \Lambda_{0\vec{Q}} \mathcal{Z} \nu_0 \bar{\nu}_{\vec{Q}}^A = 0 , \qquad (49a)$$

and

$$-\tau_Q^{-1}\bar{\nu}_Q^A + g_0\Lambda_{0\vec{Q}}\mathcal{Z}\bar{\nu}_0^A\bar{\nu}_{\vec{Q}}^A = 0 \ , \eqno(49b)$$



Figure 2. Idem as in figure 1 for two other values of parameters @.

where

$$\mathcal{Z} = e^{\beta \hbar \Delta_{0Q}} - 1 . \tag{50}$$

From Eq. (49b) we find the asymptotic saturation value of ν_0 , namely

$$\bar{\nu}_0^A = 1/g_0 \Lambda_{0Q} \mathcal{Z} \tau_Q \quad , \tag{51}$$

and replacing this value in Eq. (49a) we find that asymptotically

$$\bar{\nu}_Q^A = (\alpha_0 / \alpha_Q) \tau_Q [I_0 - I_0^*] , \qquad (52)$$

where we have introduced the quantity

$$I_0^* = [g_0 \Lambda_{0Q} \mathcal{Z} \tau_0 \tau_Q]^{-1} .$$
 (53)

Eq. (51) clearly tells us that the saturated value of ν_0 is smaller when larger the number of externally pumped modes (g_0) , when larger the relaxation time of the low frequency modes (τ_0) , and when larger the bilinear coupling coefficient (Λ_{oQ}) , as should be expected. On the other hand, Eq. (52), tells us that the population of the "exploding" mode grows linearly with the power of the external source pumping the higher frequency modes, with a proportionality factor depending on the ratio of the number of externally pumped modes to the number of the lowest frequency modes, hence a very large number. Further, the term in the square bracket indicates that the critical pumping intensity needs be much larger than the values I_0^* of Eq. (53), evidently larger and larger when smaller and smaller is α_0 which, we recall, is a measurement of the fraction of modes in the Brillouin zone pumped by the external source.

Let us now look for the asymptotic value of the quasi-chemical potential μ_Q . One has

$$\mu_Q = \hbar \omega_Q - \beta^{-1} \ln \left(\frac{1}{\nu_Q}\right) \simeq \hbar \omega_Q - (\beta \nu_Q)^{-1} , \quad (54)$$

and using Eq. (52) we obtain that

$$\mu_Q \simeq \hbar \omega_Q - kT[(\alpha_Q/\alpha_0)\tau_Q(I_0 - I_0^*)]^{-1}$$
. (55)

This last equation demon'strates, as the curve of Fig. 3 shows, that the quasi-chemical potential of the lowest frequency mode tends asymptotically to the energy of the mode as I_0 tends to infinity, but, otherwise, remains slightly below that value for I_0 larger than the critical value. Such difference is, for $I_0 \gg I_0^*$, roughly $kT\alpha_Q/\alpha_0\tau_Q I_0$, that goes to zero with increasing I_0 .



Figure 3. The dependence on the intensity of the source of the quasi-chemical potential of the lowest frequency modes.

Consequently, such very small value implies in the resulting complex behavior consisting in the occurrence

of a near Bose-Einstein-like condensation in a nonequilibrium dissipative macrostate of the system of vibrational modes, whose dynamics is governed by appropriate nonlinear laws.

V. Summary and concluding remarks

We have analized 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^[8]. High order relaxation effects - up to fourtli order in the interaction strengths - were introduced, what produced the nonlinear contributions that are shown to be relevant for the emergence of complex behavior in the system.

We have explicitly obtained the equations of evolutions for the populations of tlie vibrational modes, being able to show that a particular bilinear term can produce a remarkable effect of transferring, in a cascadelike process, the energy the different modes are receiving to the mode with the lowest frequency. In a formal writing a Bose-Einstein-lilte distribution is introduced for the population of the vibrational modes, characterized by the temperature of the bath and a quasichemical potential for each mode. The latter is zero at equilibrium (absence of the external source), as it should to produce the well known Planck distribution, but becomes non-vanishing and increasing with increasing source power. Hence, the one for the mode of lowest frequency may approach, and eventually coincide, with this frequency, leading to an "explosion" in population of such mode.

A numerical solution for a model system is described in Section IV. In fact we were able to show that, there exists a critical value of the pumping power beyond which an enormous increase in the population of the lowest frequency modes is produced, at the expenses of all the other modes. As shown, the quasi-chemical potentials of the lowest frequency modes tend only asymptotically (source intensity going to infinity) to coincide with the value of the modes' frequencies. However, the populations of the lowest frequency modes increase enormously, while those of the all 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 lowest energy state. 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 has been discussed in Section IV. Here we only emphasize that this unexpected complex behavior arises as a result of the nonlinear characteristics of the kinetic equations in tlie nonequilibrium dissipative state of the system. As indicated in the Introduction we call the phenomenon *Fröhlich effect* after Fröhlich suggestions^[12], of which ours is a detailed calculation invoking high order relaxation effects.

Concerning real systems where the situation here described may be present, we can mention two cases. One is that of polar semiconductors where there are higli frequency optical modes and low-frequency acoustic modes. We have considered the case when the optical modes are excited through the indirect photon carrier absorption process in doped materials^[23]. It is possible to show that the Frohlich effect is present, but at such too high levels of laser power that would produce extensive damage in the sample, and so is not experimentally accessible^[20]. The reason is the very low efficiency of tlie pumping procedure. Additional studies, introducing excitation by means of high intensity electric fields, are under way. Another case is that of dipolar vibrations in biopolymers and other biological material~excited by metabolic processes, and in contact with a thermal bath^[11,12]: Exact model calculations^[24] seem to show that this is a quite appropriate candidate for the actual occurrence of Frohlich effect.

Finally, we anticipate that preliminary results^[25] seem to show that the behavior of the system is even more complex, in the sense that beyond the critical intensity for the emergence of Frohlich effect, excitations in the system at the lowest frequency propagates in a coherent fashion, with almost no dissipation, and being of the solitary wave type. It is worth mentioning that the characteristics of the curve of popu-

lation vs. intensity of the pumping source (e.g. figures 1 and 2 in this paper), taking together with the single-frequency ("monochromaticism~'),and dissipationless propagation of the excitations^[25], describes a phenomenon similar to that of laser action.

Acknowlodgements

Two of the authors (ARV and RL) are Brazil National Research Council (CNPq) research fellows. We acknowledge the finantial support to our Group that through the years was provided by the State of São Paulo Research Foundation (FAPESP).

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