Eur. Phys. J. B **13**, 131–139 (2000)

THE EUROPEAN PHYSICAL JOURNAL B

EDP Sciences

Nonlinear-driven instability of dynamical plasma in solids: Emergence of spatially self-organized order and chaotic-like behavior

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Received 8 July 1998 and Received in final form 6 May 1999

Abstract. We analyze in detail the nonlinear kinetics of a carrier system in a photoinjected plasma in semiconductors under the action of constant illumination with ultraviolet light. We show that the spatially homogeneous steady-state becomes unstable, and a charge density wave emerges after a critical intensity of the incident radiation is achieved. It is shown that this instability can only follow in doped p-type materials. In bulk systems the critical intensity was found to be too high making the phenomenon not observable under realistic experimental conditions. However, a more efficient electron excitation can be obtained in low dimensional p-type systems, like some molecular and biological polymers, where the interaction may follow by chemical interaction with the medium. We show that for intensities beyond the critical threshold an increasing number of modes provide further contributions (subharmonics) to the space inhomogeneity. It is conjectured that this process could lead the system to display chaotic-like behavior.

PACS. 47.20.Hw Morphological instability; phase changes – 47.20.Ky Nonlinearity (including bifurcation theory)

1 Introduction

The presence of nonlinear terms in the kinetic equations that govern the evolution of the macroscopic state of open systems, is known to be a fundamental condition for the so-called complex behavior to arise. One of the manifestations of this behavior is the phenomenon of synergetic self-organization in dissipative (open) systems [1]. These systems, when kept under constant excitation leading to Non-equilibrium steady-state conditions, can display macroscopic spatial structures. We consider here a phenomenon of this type. Extensive and comprehensive reviews on this subject can be found in reference [2]. In this communication we report an in depth study on the possibility of spatial self-organization of a highly excited electron system. Theoretical studies have recently suggested the possibility of emergence of a morphological transition in carrier systems in bulk matter when under the action of an external pumping source of energy [3]. In this paper we focus mainly on the case of a photoinjected plasma in bulk (three-dimensional) polar semiconductors while being continuously illuminated with ultraviolet radiation. We also present some considerations involving the case of low-dimensional electron systems like biopolymers under dark-biochemical excitation.

2 Instability in a photoinjected plasma in polar semi-conductors

Let us consider a direct-gap, polar semiconductor (GaAs for instance) with energy gap $E_{\rm G}$, in contact with a thermal bath at temperature $T_{\rm B}$. The sample is driven and maintained far away from equilibrium by pumping energy with a source of UV-light. We assume that the source radiates in a broad spectrum of energies with a spectral density, $\mathfrak{g}(\hbar\omega)$, of the form

$$\mathfrak{g}(\hbar\omega) = \mathfrak{g}_0\theta(\hbar\omega - E_{\rm m})\theta(E_{\rm M} - \hbar\omega), \qquad (1)$$

where $E_{\rm m}$ and $E_{\rm M}$ are a minimum and a maximum cut-off in energy, $\mathfrak{g}_0$ is a constant and $\hbar\omega$ is the photon energy; $\theta(x)$ is the Heaviside step function. Equation (1) implies in a constant illumination with photons in the energy interval $E_{\rm m} \leq \hbar\omega \leq E_{\rm M}$, and we consider the case where $E_{\rm m} < E_{\rm G}$ and $E_{\rm M} > E_{\rm G}$ and therefore electrons in the valence band are excited to the conduction band. In this process electron-hole pairs (to be denominated carriers in what follows) with density $n(t)$ (in units of cm^{-3}) are created. When the source is turned on, a nonequilibrium carrier distribution is established in the system

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and, after a transient that typically extends in a ten-fold pioseconds scale [4], a steady-state sets in. Note that only photons with energies E in the interval $E_{\rm G} \leq E \leq E_{\rm M}$ are absorbed in the sample and then the total intensity I (in units of erg cm^{-2}s^{-1}) that effectively contributes to the absorption process is given by $I = \mathfrak{g}_0 \Delta E$, where $\Delta E = E_{\rm M} - E_{\rm G}$. The steady-state, determined only by I (once ΔE and $T_{\rm B}$ are kept fixed), is characterized by a spatial electron density distribution $n(\mathbf{r})$. At low radiation intensities, when the system is in the so-called linear regime near equilibrium, the homogeneous state ($n(\mathbf{r}) =$ const.) is expected to be stable (predominance of thermal chaos: collisions, mainly *via* the long-range Coulomb interaction, rapidly sweep away the inhomogeneities). We analyze here the conditions under which this state may become unstable against the emergence of a macroscopic spatial structure in the carrier system.

The electron system is treated in the electron-hole representation. We consider that the total carrier density, $n = V^{-1} \int n(\mathbf{r}) {\rm d}^3\mathbf{r}$ (V is the volume of the sample), is high enough for the system to be on the metallic side of the Mott transition (what typically occurs for densities of the order or higher than $n \sim 10^{16}$ cm^{-3}). Under this condition the carriers form a double photoinjected plasma, that is, the gas of excitons is almost completely ionized. The carrier system relaxes its energy in excess of equilibrium through mainly two mechanisms: radiative recombination and interaction with the lattice modes. In polar semiconductors, as a general rule, we can disregard the interaction of the carriers with the acoustic modes and deformation potential interaction with the optical phonons, only retaining Fröhlich interaction between carriers and LO-phonons. Higher order processes such as self-absorption, non-radiative recombination, Auger effect, etc., are also neglected since, in the conditions to be analyzed, they have much smaller contributions than the leading spontaneous recombination.

Let us consider the evolution equation of the electron density, $n(\mathbf{r}, t)$, at position $\mathbf{r}$ and time t. This quantity is given by the average, over the nonequilibrium ensemble which characterizes the dissipative macrostate of the system, of the *single particle density operator* $\psi^\dagger(\mathbf{r})\psi(\mathbf{r})$ at position $\mathbf{r}$, *i.e.*,

$$n(\mathbf{r}, t) = {\rm Tr}\{\psi^\dagger(\mathbf{r})\psi(\mathbf{r})\rho(t)\}, \tag{2}$$

where $\rho(t)$ is the *probability distribution operator* at time t corresponding to the nonequilibrium statistical ensemble formalism to be used, and $\psi^\dagger$ and ψ are the usual single-particle field operators. The statistical operator $\rho(t)$, to be specified below, is built on the basis of the so-called Non-Equilibrium Statistical Operator method (NESOM for short). NESOM provides mechanical-statistical basis for the construction of $\rho(t)$, the Non-Equilibrium Statistical Operator. We notice that NESOM is a particular nonequilibrium ensemble formalism founded on the ideas set forth by Gibbs and Boltzmann [5,6]. In far-from-equilibrium systems the method has been successfully applied to several experimental situations, especially in the area of photoexcited semiconductors [7], as the one we are considering here. We will not describe here the method which is presented in the books in reference [5], and the review articles in reference [6]; a description along with applications to the study of ultrafast relaxation processes in the photoinjected plasma in bulk matter is given in reference [8], while the case of polymers and quasi-onedimensional electron systems (quantum wires) is presented in references [9,10], respectively.

Expressing the field operators $\psi^\dagger(\mathbf{r})$ and $\psi(\mathbf{r})$ on the basis of single-electron creation (annihilation) operators $c^\dagger_{\mathbf{k}}(c_{\mathbf{k}})$ and $h^\dagger_{-\mathbf{k}}(h_{-\mathbf{k}})$ for electrons and holes respectively, we write equation (2) in the form

$$n(\mathbf{r}, t) = \sum_{\mathbf{Q}} n(\mathbf{Q}, t) {\rm e}^{-{\rm i}\mathbf{Q}.\mathbf{r}}, \tag{3}$$

where the Fourier transform, $n(\mathbf{Q}, t)$, of the carrier density is given by

$$n(\mathbf{Q}, t) = \sum_{\mathbf{k}} {\rm Tr}\{c^\dagger_{\mathbf{k}+\mathbf{Q}} c_{\mathbf{k}} \rho(t)\} + \sum_{\mathbf{k}} {\rm Tr}\{h_{-\mathbf{k}-\mathbf{Q}} h^\dagger_{-\mathbf{k}} \rho(t)\}. \tag{4}$$

Wavevectors $\mathbf{Q}$ and $\mathbf{k}$ in the sums in equations (3, 4) run over the Brillouin zone (the spin index has been omitted), and Bloch's wavefunctions have been approximated by plane waves. Next we define the variables

$$n^{\rm e}_{\mathbf{k},\mathbf{Q}}(t) = {\rm Tr}\{c^\dagger_{\mathbf{k}+\mathbf{Q}} c_{\mathbf{k}} \rho(t)\}, \tag{5}$$

$$n^{\rm h}_{\mathbf{k},\mathbf{Q}}(t) = {\rm Tr}\{h_{-\mathbf{k}-\mathbf{Q}} h^\dagger_{-\mathbf{k}} \rho(t)\}, \tag{6}$$

which are the average values, over the nonequilibrium ensemble, of the Dirac-Wigner-Landau single-particle dynamical operators, in this case for electrons and for holes respectively, and once the transport equations for $n^{\rm e}_{\mathbf{k},\mathbf{Q}}(t)$ and $n^{\rm h}_{\mathbf{k},\mathbf{Q}}(t)$ are obtained, the corresponding one for $n(\mathbf{r}, t)$ follows from equations (3, 4).

For the description of the nonequilibrium statistical thermodynamics of this photoinjected double plasma (dealt with, we recall, in the single-particle description) first we take, as basic variables, the total carrier energy, $E_{\rm c}(t)$, and electron and hole densities, $n_{\rm e}(t)$ and $n_{\rm h}(t)$, that is (see Ref. [11])

$$E_{\rm c}(t) = {\rm Tr}\{\widehat{H}_{\rm c} \rho(t)\}, \tag{7}$$

$$n_{\rm e(h)}(t) = \frac{1}{V} {\rm Tr}\{\widehat{N}_{\rm e(h)} \rho(t)\}, \tag{8}$$

where

$$\widehat{H}_{\rm c} = \sum_{\mathbf{k}} \left\{ \varepsilon^{\rm e}_{\mathbf{k}} c^\dagger_{\mathbf{k}} c_{\mathbf{k}} + \varepsilon^{\rm h}_{\mathbf{k}} h^\dagger_{\mathbf{k}} h_{\mathbf{k}} \right\} \tag{9}$$

is the carriers' Hamiltonian, and

$$\widehat{N}_{\rm e} = \sum_{\mathbf{k}} c^\dagger_{\mathbf{k}} c_{\mathbf{k}}; \qquad \widehat{N}_{\rm h} = \sum_{\mathbf{k}} h^\dagger_{\mathbf{k}} h_{\mathbf{k}} \tag{10}$$

are the electron and hole number operator. Second, for dealing with the local spatial characteristics of the electron systems, we must also include the variables of equations (5, 6). In equation (9) $\varepsilon^{\rm e}_{\mathbf{k}} = E_{\rm G} + (\hbar^2/2m_{\rm e})k^2$ and

$\varepsilon^{\rm h}_{\mathbf{k}} = (\hbar^2/2m_{\rm h})k^2$ are the band energies of electrons and holes respectively, in the effective mass approximation ($m_{\rm e(h)}$ denotes the electron (hole) effective mass).

Consequently, according to NESOM, the basic set of dynamical variables, on which the nonequilibrium distribution depends, consists of

$$\Big\{\widehat{H}_{\rm c}, \widehat{N}_{\rm e}, \widehat{N}_{\rm h}, c^\dagger_{\mathbf{k+Q}}c_{\mathbf{k}}, h_{-\mathbf{k}-\mathbf{Q}}h^\dagger_{-\mathbf{k}}\Big\}, \tag{11}$$

with $\mathbf{Q} \neq 0$.

The NESOM-nonequilibrium statistical operator $\rho(t)$ introduced in equation (2) is a superoperator built in terms of the basic dynamical variables of equation (11) which, in Zubarev's approach [6], is given by

$$\rho(t) = \exp\left\{-\widehat{S}(t,0) + \int_{-\infty}^{t} \mathrm{e}^{\varepsilon(t-t)}\frac{\mathrm{d}}{\mathrm{d}t'}\widehat{S}(t', t'-t)\mathrm{d}t'\right\}, \tag{12}$$

where

$$\begin{aligned}\widehat{S}(t,0) = -\log\bar{\rho}(t,0) = \\ \phi(t) + \beta_{\rm c}(t)\Big[\widehat{H}_{\rm c} - \mu_{\rm e}(t)\widehat{N}_{\rm e} - \mu_{\rm h}(t)\widehat{N}_{\rm h}\Big] \\ + \sum_{\mathbf{k},\mathbf{Q}}\Big[F^{\rm e}_{\mathbf{kQ}}(t)c^\dagger_{\mathbf{k+Q}}c_{\mathbf{k}} + F^{\rm h}_{\mathbf{kQ}}(t)h_{-\mathbf{k}-\mathbf{Q}}h^\dagger_{-\mathbf{k}}\Big]\end{aligned} \tag{13}$$

is the so-called *informational entropy operator* [12], and where $\beta_{\rm c}(t), -\mu_{\rm e}(t)\beta_{\rm c}(t), -\mu_{\rm h}(t)\beta_{\rm c}(t)$, $F^{\rm e}_{\mathbf{kQ}}(t)$ and $F^{\rm h}_{\mathbf{kQ}}(t)$ are the Lagrange multipliers (intensive nonequilibrium thermodynamic variables) that the method introduces. The first three ones are those associated to the homogeneous variables $E_{\rm c}(t)$, $N_{\rm e}(t)$ and $N_{\rm h}(t)$, and $\beta_{\rm c}$ is usually written as $\beta_{\rm c}(t) = 1/k_{\rm B}T^*_{\rm c}(t)$ introducing, in this way, the so-called quasitemperature $T^*_{\rm c}(t)$ for the carrier system; quantities $\mu_{\rm e}(t)$ and $\mu_{\rm h}(t)$ are the so-called quasi-chemical potentials for electrons and for holes respectively [13]. The other Lagrange multipliers are related to the Dirac-Wigner-Landau dynamical single-particle operators. In equation (13) $\bar{\rho}(t,0)$ is the auxiliary statistical operator

$$\bar{\rho}(t,0) = \exp\Big\{\widehat{S}(t,0)\Big\}, \tag{14}$$

sometimes called the coarse-grained part of the fine-grained statistical operator of equation (12), or distribution for a "frozen" instantaneous quasi-equilibrium [6,7], which has a relevant role in the theory and provides the foundations for a statistical irreversible thermodynamics [14]. The Lagrange multiplier $\phi(t)$ (playing the role of the logarithm of a nonequilibrium partition function) ensures the normalization of the statistical operator. The informational entropy operator in Heisenberg representation, which appears in equation (12), is given by

$$\widehat{S}(t', t'-t) = \exp\left\{-\frac{1}{\mathrm{i}\hbar}(t'-t)\widehat{H}\right\}\widehat{S}(t,0)\exp\left\{\frac{1}{\mathrm{i}\hbar}(t'-t)\widehat{H}\right\}. \tag{15}$$

The quantity ε (> 0) in equation (12) is an infinitesimal that goes to zero after the trace operation in the calculation of averages has been performed. As expected, after switching the external perturbation off, the statistical operator of equation (12) converges to the grand-canonical distribution in equilibrium [15].

We derive next the evolution equations for the basic macrovariables of equations (5–8) resorting to the NESOM-based nonlinear quantum kinetic theory [6,16], but restricted to the Markovian approximation (also dubbed second order approximation in relaxation theory) [17], and which can be considered as a far-reaching generalization of Mori-Heisenberg-Langevin equations (see for example Ref. [18]). The transport equations for $E_{\rm c}(t)$ and $n(t)$ are given, for example, in reference [11], and those for $n^{\rm e(h)}_{\mathbf{k},\mathbf{Q}}(t)$ are (see Refs. [10,19])

$$\begin{aligned}\frac{\partial}{\partial t}n^{\rm e}_{\mathbf{k},\mathbf{Q}}(t) = -\frac{1}{\mathrm{i}\hbar}(\varepsilon^{\rm e}_{\mathbf{k+Q}} - \varepsilon^{\rm e}_{\mathbf{k}})n^{\rm e}_{\mathbf{k},\mathbf{Q}}(t) \\ + \frac{1}{i\hbar}\mathcal{V}(\mathbf{Q})\left[f^{\rm e}_{\mathbf{k+Q}}(t) - f^{\rm e}_{\mathbf{k}}(t)\right]n(\mathbf{Q},t) - A^{\rm e}_{\mathbf{k},\mathbf{Q}}(t)n^{\rm e}_{\mathbf{k},\mathbf{Q}}(t) \\ + A^{\rm h}_{\mathbf{k},\mathbf{Q}}(t)n^{\rm h}_{\mathbf{k},\mathbf{Q}}(t) + \mathcal{L}^{\rm e}_{\mathbf{k},\mathbf{Q}}(t) + \mathcal{N}^{\rm e}_{\mathbf{k},\mathbf{Q}}(t),\end{aligned} \tag{16}$$

$$\begin{aligned}\frac{\partial}{\partial t}n^{\rm h}_{\mathbf{k},\mathbf{Q}}(t) = \frac{1}{\mathrm{i}\hbar}(\varepsilon^{\rm h}_{\mathbf{k+Q}} - \varepsilon^{\rm h}_{\mathbf{k}})n^{\rm h}_{\mathbf{k},\mathbf{Q}}(t) \\ - \frac{1}{\mathrm{i}\hbar}\mathcal{V}(\mathbf{Q})[f^{\rm h}_{\mathbf{k+Q}}(t) - f^{\rm h}_{\mathbf{k}}(t)]n(\mathbf{Q},t) \\ - A^{\rm h}_{\mathbf{k},\mathbf{Q}}(t)n^{\rm e}_{\mathbf{k},\mathbf{Q}}(t) + A^{\rm e}_{\mathbf{k},\mathbf{Q}}(t)n^{\rm h}_{\mathbf{k},\mathbf{Q}}(t) \\ + \mathcal{L}^{\rm h}_{\mathbf{k},\mathbf{Q}}(t) + \mathcal{N}^{\rm h}_{\mathbf{k},\mathbf{Q}}(t),\end{aligned} \tag{17}$$

where the quantities $f^{\rm e(h)}_{\mathbf{k}}(t)$ are

$$f^{\rm e}_{\mathbf{k}}(t) = \mathrm{Tr}\Big\{c^\dagger_{\mathbf{k}}c_{\mathbf{k}}\rho(t)\Big\}, \tag{18a}$$

$$f^{\rm h}_{\mathbf{k}}(t) = \mathrm{Tr}\Big\{h^\dagger_{\mathbf{k}}h_{\mathbf{k}}\rho(t)\Big\}, \tag{18b}$$

that is, they are the populations in state $|\mathbf{k}\rangle$ in the nonequilibrium ensemble. In equations (16,17) $\mathcal{V}(\mathbf{Q})$ is the matrix element of the Coulomb interaction between electrons dealt with in RPA (in bulk matter this potential is $\mathcal{V}(\mathbf{Q}) = 4\pi e^2/V\epsilon_0 Q^2$, where $Q \equiv |\mathbf{Q}|$ and ϵ_0 is the background dielectric constant); $\mathcal{N}^{\rm e(h)}_{k,Q}$ are bilinear contributions in $n^{\rm e(h)}_{\mathbf{kQ}}(t)$ whose cumbersome expressions we omit to write down for brevity since they will not be present in the linear stability analysis to be performed; terms $\mathcal{L}^{\rm e(h)}_{\mathbf{k},\mathbf{Q}}(t)$ are responsible for the electron-LO-phonon interaction (which are proportional to the square modulus of the matrix element of Fröhlich potential).

As it can be numerically demonstrated (we used parameters characteristic of GaAs semiconductor, but this result is expected to be general for any other direct-gap polar semiconductor) the contributions $\mathcal{L}$ can be disregarded (in comparison with the other linear terms in Eqs. (16,17)), for wavenumber Q not too close to the zone center (in GaAs, for example, neglecting $\mathcal{L}$ is quite satisfactory for all $Q \gtrsim 10^2\,\mathrm{cm}^{-1}$). We shall see, as we proceed,

that the instability of the homogeneous state (first bifurcation) arises for values of Q near the Brillouin wavenumber $Q_{\rm B}$ in (GaAs $Q_{\rm B} \sim 5 \times 10^7$ cm^{-1}) and therefore the terms $\mathcal{L}^{\rm e(h)}_{\mathbf{k},\mathbf{Q}}(t)$ can be, in effect, disregarded. Terms $A^{\rm e(h)}_{\mathbf{k},\mathbf{Q}}$ are responsible for the interaction between carriers and the radiation fields, and are given by

$$A^{\rm e(h)}_{\mathbf{k},\mathbf{Q}}(t) = \mathfrak{s}_R(\varepsilon^{\rm e}_{\mathbf{k}} + \varepsilon^{\rm h}_{\mathbf{k}}) f^{\rm e(h)}_{\mathbf{k}}(t) + \mathfrak{s}_F \frac{\mathfrak{g}(\hbar\omega_{\mathbf{k}})}{(\varepsilon^{\rm e}_{\mathbf{k}} + \varepsilon^{\rm h}_{\mathbf{k}})^2} + (\text{same} \quad \text{with} \quad \mathbf{k} \to \mathbf{k} + \mathbf{Q}), \tag{19}$$

where $\mathfrak{g}(\hbar\omega_{\mathbf{k}})$ is the spectral density defined in equation (1) evaluated at the energy $\hbar\omega_{\mathbf{k}} = \varepsilon^{\rm e}_{\mathbf{k}} + \varepsilon^{\rm h}_{\mathbf{k}}$, and $\mathfrak{s}_R$ and $\mathfrak{s}_F$ are the constants given by

$$\mathfrak{s}_R = \frac{\eta_\infty e^2 |\mathbf{P}_{\rm vc}|^2}{\hbar^2 m_0^2 c^3}, \qquad \mathfrak{s}_F = \frac{2\pi^2 \hbar e^2 |\mathbf{P}_{\rm vc}|^2}{\eta_\infty m_0^2 c}, \tag{20}$$

where e is the electron charge, $|\mathbf{P}_{\rm vc}|^2$ the square modulus of the matrix of the electron linear momentum between conduction and valence bands states at the zone center, m_0 the electron rest mass, c the speed of light and η_∞ the high frequency refraction index. Terms $A^{\rm e(h)}_{\mathbf{k},\mathbf{Q}}(t)$ are composed of two contributions, one associated to the recombination processes, namely, the first term on the right hand side of equation (19) plus the corresponding one with $\mathbf{k} \to \mathbf{k} + \mathbf{Q}$, and another one associated to the interaction between carriers and the external radiation field, namely, the second on the right of equation (19) plus the corresponding one with $\mathbf{k} \to \mathbf{k} + \mathbf{Q}$.

The homogeneous steady state is characterized by the $\mathbf{Q} = 0$ Fourier amplitude in equation (3) and null variables $n^{\rm e(h)}_{\mathbf{k},\mathbf{Q}}$ (defined in Eqs. (5, 6) for $\mathbf{Q} \neq 0$). We are next going to analyze a possible instability of the stationary homogeneous state against the formation of a spatial pattern, *i.e.*, when quantities $n^{\rm e(h)}_{\mathbf{k},\mathbf{Q}}$ become different from zero (for $\mathbf{Q} \neq 0$). For this purpose we consider the stability of the homogeneous state resorting to a linear stability analysis. Recalling that in the homogeneous state $n^{\rm e(h)}_{\mathbf{k},\mathbf{Q}} = 0$, and then $\mathcal{N}^{\rm e(h)}_{k,Q}$ in equations (16, 17) are null, we test the evolution of the system after we impose an arbitrary small perturbation of the form

$$n^{\rm e(h)}_{\mathbf{k},\mathbf{Q}}(t) = n^{\rm e(h)}_{\mathbf{k},\mathbf{Q}}(0) \exp\lambda t, \tag{21}$$

where $\lambda = \gamma + {\rm i}\omega$, and we analyze the sign of γ. At low radiation intensities γ is negative for all $\mathbf{Q}$ in the Brillouin zone, and then the perturbation decays and the homogeneous state remains stable. If an instability arises against a sinoidal structure of wavevector $\mathbf{Q} \neq 0$ at higher levels of excitation, then γ must be equal to zero at some critical intensity I_c, and change sign thereafter. Therefore, taking $\gamma = 0$ in equation (21), introducing it in equations (16, 17), neglecting the bilinear terms $\mathcal{N}^{\rm e(h)}_{k,Q}(t)$ and performing a sum over $\mathbf{k}$, we arrive to the equation that determines the steady-state solutions $n(\mathbf{Q})$ at the critical intensity. This equation is given by

$$n(\mathbf{Q})\epsilon(\mathbf{Q},\omega) = 0, \tag{22}$$

where

$$\epsilon(\mathbf{Q},\omega) = 1 - \mathcal{V}(\mathbf{Q}) \sum_{\mathbf{k}} \frac{j_1(\mathbf{k},\mathbf{Q},\omega) + {\rm i} j_2(\mathbf{k},\mathbf{Q},\omega)}{j_3(\mathbf{k},\mathbf{Q},\omega) + {\rm i} j_4(\mathbf{k},\mathbf{Q},\omega)} \tag{23}$$

and

$$j_1(\mathbf{k},\mathbf{Q},\omega) = -\frac{1}{\hbar}(f^{\rm e}_{\mathbf{k}+\mathbf{Q}} - f^{\rm e}_{\mathbf{k}})(\hbar\omega + \varepsilon^{\rm h}_{\mathbf{k}+\mathbf{Q}} - \varepsilon^{\rm h}_{\mathbf{k}}) + \frac{1}{\hbar}(f^{\rm h}_{\mathbf{k}+\mathbf{Q}} - f^{\rm h}_{\mathbf{k}})(\hbar\omega - \varepsilon^{\rm e}_{\mathbf{k}+\mathbf{Q}} + \varepsilon^{\rm e}_{\mathbf{k}}), \tag{24}$$

$$j_2(\mathbf{k},\mathbf{Q},\omega) = [(f^{\rm e}_{\mathbf{k}+\mathbf{Q}} - f^{\rm e}_{\mathbf{k}}) - (f^{\rm h}_{\mathbf{k}+\mathbf{Q}} - f^{\rm h}_{\mathbf{k}})](A^{\rm e}_{\mathbf{k},\mathbf{Q}} + A^{\rm h}_{\mathbf{k},\mathbf{Q}}), \tag{25}$$

$$j_3(\mathbf{k},\mathbf{Q},\omega) = \frac{1}{\hbar}(\hbar\omega - \varepsilon^{\rm e}_{\mathbf{k}+\mathbf{Q}} + \varepsilon^{\rm e}_{\mathbf{k}})(\hbar\omega + \epsilon^{\rm h}_{\mathbf{k}+\mathbf{Q}} - \varepsilon^{\rm h}_{\mathbf{k}}), \tag{26}$$

$$j_4(\mathbf{k},\mathbf{Q},\omega) = -(\hbar\omega + \varepsilon^{\rm h}_{\mathbf{k}+\mathbf{Q}} - \varepsilon^{\rm h}_{\mathbf{k}})A^{\rm h}_{\mathbf{k},\mathbf{Q}} - (\hbar\omega - \varepsilon^{\rm e}_{\mathbf{k}+\mathbf{Q}} + \varepsilon^{\rm e}_{\mathbf{k}})A^{\rm e}_{\mathbf{k},\mathbf{Q}}. \tag{27}$$

In equations (24, 27) the populations $f^{\rm e(h)}_{\mathbf{k}}$ (defined in Eqs.(18a, 18b)), after linearization of equations (16, 17), acquire in the homogeneous state of reference a form reminescent of Fermi-Dirac distributions. For a discussion of this point see for example references [11,12].

We stress that $\epsilon(\mathbf{Q},\omega)$ defined in equation (23) is the frequency- and wavevector-dependent *dielectric function* of the system, in the given *nonequilibrium conditions*. As known, the dielectric function has information on all the optical properties and elementary excitations (single- particles and collective modes) of the system [20]. The possibility of recognizing a physical meaning for the function $\epsilon(\mathbf{Q},\omega)$ is, certainly, of relevance. In Section 3 we shall show that, on the basis of physical arguments, the succession of instabilities arising beyond the first bifurcation can be characterized, a task that, otherwise, requires a hard numerical work.

Equation (22) admits two types of solutions, one is $n(\mathbf{Q}) = 0$ which is the solution corresponding to the homogeneous state, and another one is $n(\mathbf{Q}) \neq 0$, corresponding to the non-homogeneous state, which is possible when $\epsilon(\mathbf{Q},\omega) = 0$. Since $\epsilon(\mathbf{Q},\omega)$ is a complex function (of real arguments $\mathbf{Q}$ and ω) to set it equal to zero requires that both its real (Re ϵ) and imaginary (Im ϵ) parts be null. After introducing in equation (23) the expressions for $j_l(\mathbf{k},\mathbf{Q},\omega)(l = 1$ to $4)$, as given in equations (24–27) and going to the continuum limit in $\mathbf{k}$-space (and therefore the sum in equation (23) can be appropriately replaced by an integral over the Brillouin zone), we can proceed to look for the roots of the coupled set of equations Re $\epsilon(\mathbf{Q},\omega) = 0$ and Im $\epsilon(\mathbf{Q},\omega) = 0$ resorting to numerical integration which requires some careful handling.

We consider the case of a GaAs semiconductor although the results we will describe are expected to be valid

for any direct-gap polar semiconductors (we recall that $m_e \sim 0.05m_0$; $m_h \sim 0.6m_0$; $\epsilon_0 \sim 10$; $\eta_\infty \sim 3.5$; $E_G \sim 1.54$ eV, at room temperature; $|\mathbf{P}_{vc}|^2/m_0E_G \sim 7.41$; $Q_B \sim 5 \times 10^7$ cm^{-1}). Our calculations show that, for $\omega \neq 0$, there are not simultaneous roots for Re ϵ and Im ϵ and then the instability, if it exists, is stationary ($\omega = 0$). Therefore we look for possible roots of the static (but wavenumber-dependent) dielectric function $\epsilon(\mathbf{Q}, 0)$. Equation (23) tells us that $\epsilon(\mathbf{Q}, \omega)$ has definite parity: Re ϵ is odd in ω whereas Im ϵ is even in ω, and then Im $\epsilon(\mathbf{Q}, 0) = 0$, for all $\mathbf{Q}$ (having recognized that $\varepsilon(\mathbf{Q}, \omega)$ is the dielectric function of the system, its parity follows immediately from known results on the electrodynamics of material media [21]). Therefore, we proceed to look for the possible roots of the real part of the static dielectric function which is given by

$$\epsilon(\mathbf{Q}, 0) = 1 - \mathcal{V}(\mathbf{Q}) \sum_{\mathbf{k}} \frac{j_1(\mathbf{k}, \mathbf{Q}, 0) j_3(\mathbf{k}, \mathbf{Q}, 0) + j_2(\mathbf{k}, \mathbf{Q}, 0) j_4(\mathbf{k}, \mathbf{Q}, 0)}{[j_3(\mathbf{k}, \mathbf{Q}, 0)]^2 + [j_4(\mathbf{k}, \mathbf{Q}, 0)]^2} . \quad (28)$$

This function depends on the intensity I, the effective interval of absorption ΔE, the bath temperature T_B and the doping concentration n_0.

Simplified expressions for $\epsilon(\mathbf{Q}, 0)$ under different regimes of illumination were analyzed in some limiting conditions and reported in reference [3], showing that, at low intensity, no zero of $\epsilon(\mathbf{Q}, 0)$ is possible and therefore the homogeneous state is always stable. At high intensities, the sum in equation (28) was shown to be nearly independent of $\mathbf{Q}$ and a root of the real part can be obtained (first bifurcation) only in the case of doped p-type materials. We retake here the question using the full exact expression of equation (28). For fixed values of ΔE and T_B we look for the possible roots of $\epsilon(\mathbf{Q}, 0)$ in $\mathbf{Q}$-space, for different intensities I, considering intrinsic and doped semiconductors. The numerical calculations were carried out using parameters of GaAs but, as already said, the results to be derived are expected to be a common characteristic of all direct-gap polar semiconductors.

After a series of an in depth numerical analysis of the static wavenumber-dependent dielectric function, in several different conditions, two main laws appear to hold:

i) *for intrinsic and n-type-doped materials no root of* $\epsilon(\mathbf{Q}, 0)$ *is possible and, therefore, the spatially homogeneous state is stable under any conditions;*

ii) *for p-type-doped materials the static dielectric function,* $\epsilon(\mathbf{Q}, 0)$*, presents one root at a critical intensity* I_c*; this root appears for a wavenumber* Q_c *near the end of the Brillouin zone.*

At the critical intensity the homogeneous state becomes unstable against the emergence of a new structure of the form

$$n_c(\mathbf{r}) = n_c + \frac{1}{2}[n(\mathbf{Q}_c) e^{i\mathbf{Q}_c \cdot \mathbf{r}} + c.c.], \quad (29)$$

where n_c is the photoinjected density at I_c. The dependence of the density n on the intensity I, for radiation levels such that $n \sim 10^{19}$ cm^{-3}, is discussed in reference [11], and at higher intensities the system reaches optical saturation, for $n \sim 10^{20}$ cm^{-3}, when n becomes a constant independent of I.

We consider a p-type GaAs sample, with p-doping, density $n_0 = 6 \times 10^{18}$ cm^{-3}, bath temperature $T_B = 300$ K and illumination provided by an UV-light source with $E_m < E_G$ and $E_M = 1.7$ eV (see Eq. (1)) and hence $\Delta E \sim 200$ meV. Under these experimental conditions we find that the critical wavenumber is $Q_c \cong 1.32 \times 10^7$ cm^{-1} meaning that the spatial pattern is a sinoidal structure with wavelength $\lambda \cong 50$ Å, which is roughly larger than ten times the length of the elementary crystal cell, $a \sim 4.5$ Å. The critical intensity is found to be $I_c \cong 0.85$ TWcm^{-2}. This is a very large value which, even if available in a laboratory (maybe resorting to UV synchrotron radiation), would produce crippling material damage (if not destruction) in the sample. Consequently, the phenomenon we have evidenced, even though theoretically possible, cannot be accomplished in bulk semiconductors under realistic experimental conditions. The main reason for this fact is that the process of radiation absorption is very inefficient in these systems.

3 Morphological instability in low-dimensional systems

The theoretical results of the previous section, indicating the possibility of complex behavior in the electron plasma, lead us to consider other type of systems where the phenomenon could be observable under feasible laboratory conditions. This is the case of low-dimensional systems, which show peculiar characteristics not present in bulk matter [22]. An analysis, based on NESOM, of the optical and dynamical properties of a far-from-equilibrium quasi-one-dimensional electron system is reported in reference [10]. Particular and quite interesting systems are molecular [23] or biological [24] polymers. Instead of using UV-light illumination these materials can also be excited *via* efficient metabolic biochemical mechanisms [25]. Moreover, biopolymers may behave as a kind of semiconductor-like materials and, in particular, are of the p-doped type [23,24]. This is a quite interesting point in view of the results presented in Section 2 and of considerations advanced by Szent-György (see for example Ref. [26]). Since biochemical processes provide an alternative mechanism for electron excitation, accessible power levels could then lead to the emergence of the morphological transition. For this reason it is tempting to proceed with an analysis of the behavior of the system beyond the first bifurcation point. We recall that, at this first instability of the homogeneous state a steady-state charge density wave (SSCDW) sets in [*cf* Eq. (29)] but an analysis of the dielectric function $\epsilon(\mathbf{Q}, \omega)$ allows us to show that when increasing the radiation intensity I beyond I_c further instabilities do appear.

For $I > I_c$ we would need to analyze $\epsilon(\mathbf{Q}, 0)$ after the new inhomogeneous state $n_c(\mathbf{r})$ [*cf.* Eq (29)] has been stabilized in the sample. In this case, the whole set of

equations (16, 17) containing the nonlinear contributions need to be considered and a new stability analysis of the inhomogeneous state of reference $n_c(\mathbf{r})$ performed. However, the rigourous mathematical treatment of the resulting equations becomes quite difficult and the analysis needs to be simplified. In effect, by noting that the nonlinear terms in the amplitudes $n(\mathbf{Q})$ contribute quadratically in $\epsilon(\mathbf{Q},0)$ and are very small ($|n(\mathbf{Q}_c)|^2 \ll n_c$) for intensities near (but larger) than I_c (see Ref. [3]), they can be neglected in a first approximation. Thus, we can analyze $\epsilon(\mathbf{Q},0)$ *vs.* $I(\gtrsim I_c)$ taking its expression in the homogeneous state. Such analysis shows that for any intensity I higher than $I_c, \epsilon(\mathbf{Q},0)$ presents two roots, one at wavenumber $Q_m < Q_c$ and other one at $Q_M > Q_c$ (at the critical intensity I_c these three values coincide, *i.e.*, $Q_m = Q_c = Q_M$) (see insert in Fig. 1). For a given (fixed) intensity $I' > I_c$, the dielectric function $\epsilon(\mathbf{Q},0)$ is negative in the corresponding interval $Q'_m < Q < Q'_M$, which defines a portion of the Brillouin zone of unstable modes. This result allows us to characterize, on the basis of physical arguments, the instability and the new structures emerging out of a succession of critical points: in effect, the quantity $\epsilon(\mathbf{Q},0)$, being the static dielectric function of the system, must always be a positive real number, as should also be the refractive index which is the square root of ϵ (we recall that the imaginary part of $\epsilon(\mathbf{Q},\omega)$ is null for $\omega = 0$). Therefore, null or negative values of the static dielectric function point an unphysical behavior of the system for intensities larger than I_c. As we have seen, for a pumping intensity I_c, the static dielectric function becomes zero for $Q = Q_c$ and, for all I in the interval $I_c < I < I'$, it becomes negative for wavenumbers Q in the interval $Q'_m < Q < Q'_M$. This implies that, for increasing values of I beyond I_c, $\epsilon(\mathbf{Q},0)$ goes through zeroes for all wavenumbers in such interval. Therefore, for those values of the wavenumber, the so-called dielectric response $\epsilon^{-1}(\mathbf{Q},0)$ becomes infinite, indicating an instability of the charge density at such wavenumbers, in complete analogy with the criteria of phase transitions in equilibrium (see Appendix A).

This analysis indicates that beyond I_c, when the density is $n_c(\mathbf{r})$ of equation (29), as the intensity increases a growing number of stable modes contributes to compose the spatial charge density leading, at $I'(> I_c)$, to a stable structure of the form

$$n(\mathbf{r}) \cong n_s + \sum_{Q'_m \le Q \le Q'_M} n(\mathbf{Q}) \mathrm{e}^{\mathrm{i}\mathbf{Q}.\mathbf{r}}. \qquad (30)$$

We recall that this is an approximation, since the exact expression would not be linear in the amplitudes as it is the simple superposition in this equation (30). However, once conditions leading to small amplitudes $n(\mathbf{Q})$ are present, equation (30) would constitute an acceptable approximation. Therefore, with increasing intensity I above I_c, we may expect the emergence of a charge density wave composed of a large number of normal modes.

At this point an important question needs to be considered, namely, that of imposing boundary conditions. In fact, taking into account that the sample is finite in size, if L is its length in the direction of $\mathbf{Q}$, boundary conditions require that the permissible wavenumbers are $Q_m^{(l)} = (l_m - l + 1)\pi/L$ and $Q_M^{(l)} = (l_M + l - 1)\pi/L$, where l_m and l_M are the integers corresponding to the wavenumbers at the first instability, and l are integers ($l = 1, 2, 3 \ldots$). Moreover, there are upper and lower limiting values, *i.e.*, $Q^{(l)}$ must be in the intervals such that $1 \le l \le l_m$ (for $Q_m^{(l)}$) and $1 \le l \le l_B$ (for $Q_M^{(l)}$) with l_B being the maximum integer value for which $Q_M^{(l_B)} < Q_B$. These wavenumbers have associated characteristic wavelengths that are said to be extrinsic, in the sense that they are dependent on the size (geometry) of the system [27,28].

Figure 1 shows the linear stability diagram indicating the succession of bifurcations of the homogeneous steady state. This is a diagram which resembles, among others, the case of the so-called bimolecular model (or Brussellator), the Bènard problem and, fittingly, the Turing instability against morphological ordering in reaction-diffusion systems. In fact, the succession of critical points indicating the instabilities of the homogeneous state against spatial waves with wavenumbers $Q_m^{(l)}$ and $Q_M^{(l)}$ is quite analogous to the case of Bènard and Turing instabilities [27,28].

These characteristics of a steady-state charge density wave with a complex structure (containing the linear superposition of modes as in Eq.(30) plus nonlinear corrections) lead us to the prediction of a particular asymptotic phenomenon. It consists in that the growing number of modes, contributing to the emergence of the complex spatial ordering, would lead to the presence of an excess of modes in the system in such a way that it would show what may resemble chaotic behavior. Let us call this phenomenon *turbulent-like chaos*, following Landau's [29] and Prigogine's [30] ideas who have discussed turbulence and chemical chaos, respectively, in terms of an overexcess of modes emerging in the system. We may say that, with increasing pumping power, a particular route to this chaotic-like state arises, which we term as *Landau-Prigogine route to turbulent-like chaos*, qualitatively described in Figure 2. This figure has been adapted from one in reference [30] (p. 168), but it must be noticed that here it is referring to local variations in space of the steady-state carriers' plasma. Moreover, pursuing the analogy with the Belousov-Zhabotinskii reaction it may be conjectured that, with further increasing intensity of the pumping source, new levels of complex behavior could arise in the steady-state spatial distribution of the carriers' charge density. Possible steps would be regimes of chaotic behavior accompanied with mixed-mode oscillations, chaotic and partially periodic relaxation oscillations, etc.

4 Concluding remarks

We have reported the possible emergence of spatial order in a carrier plasma when under the action of an external source of energy. We have shown that the electron system in semiconductors under continuous illumination with UV-light can display an instability

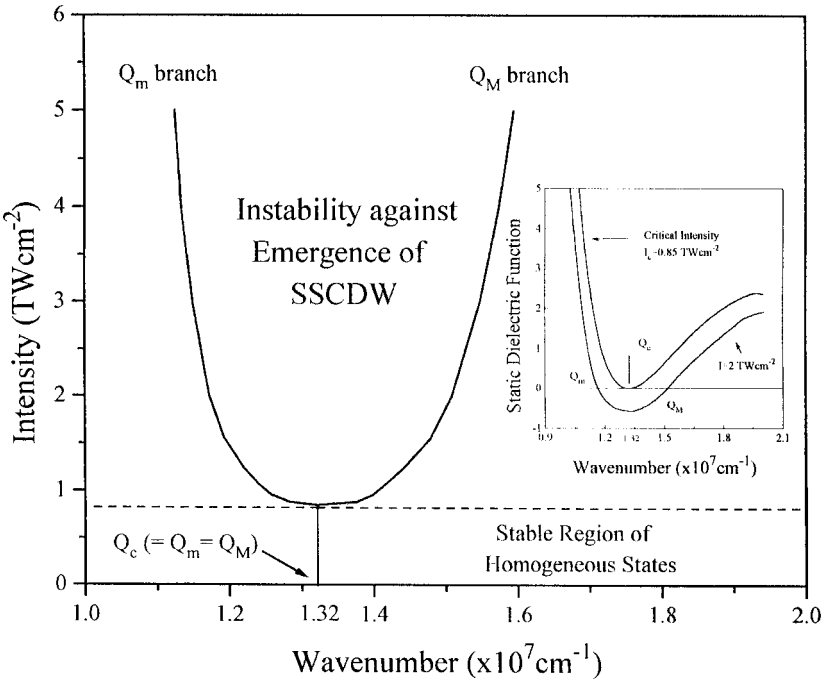

Fig. 1. Linear stability diagram (see main text); the insert shows the behavior of $\epsilon(Q,0)$ around Q_c for the critical intensity I_c and for $I > I_c$.

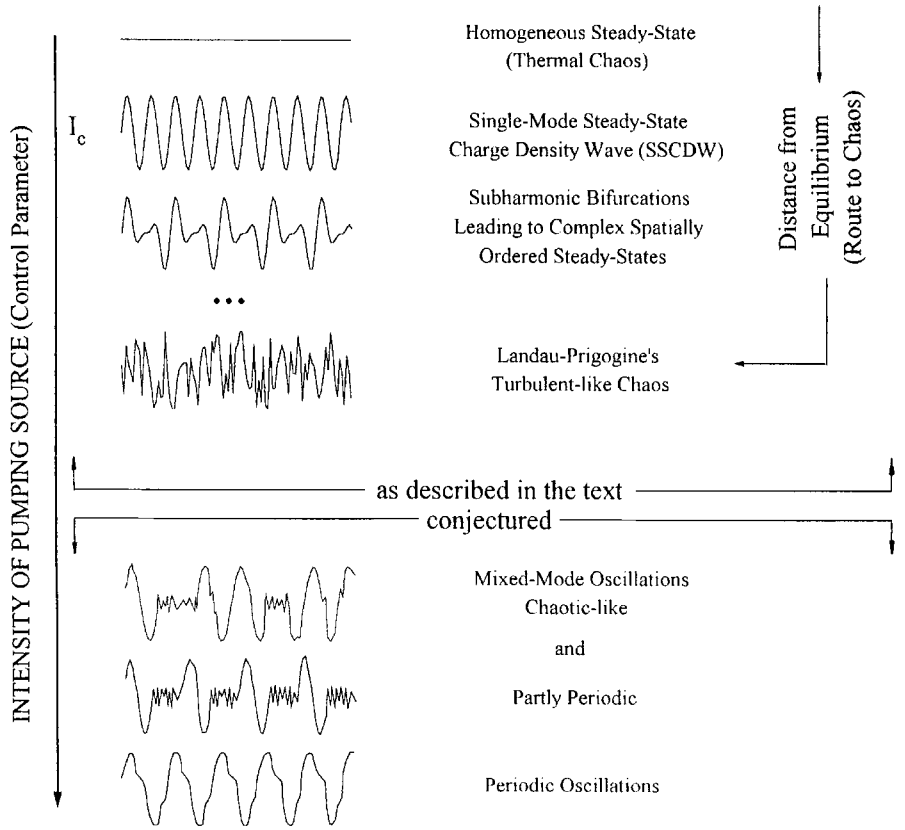

Fig. 2. Schematic qualitative description of the route to the so-called turbulent-like chaos described in the text.

of the homogeneous state against the formation of a steady-state spatial charge-density-wave structure. We have based our analysis on the mechanical-statistical formalism of NESOM, which allowed us to derive the transport equation [*cf.* Eqs.(16, 17)] of the electron density $n(\mathbf{r}, t)$ defined in equation (2). We showed numerically that the instability of the homogeneous state arises only in doped p-type materials (for intrinsic and n-doped samples the homogeneous state is stable under any intensity of the pumping source). Moreover, the critical wavenumber $Q_{\rm c}$ that characterize the spatial pattern at the first bifurcation is at near the end of the Brillouin zone. However, the critical intensity of the pumping source necessary for the phenomenon to follow was found to be so high that, even if available in a laboratory, it would produce sample damage. The high value of $I_{\rm c}$ required for the phenomenon to arise is a consequence of the low efficiency of the radiation absorption mechanism by electrons: the contribution associated to the interaction between electrons and the external radiation in equation (19) is smaller than the one corresponding to pair recombination, except at high intensities when only the bifurcation can follow.

Nevertheless, as discussed in Section 3, materials with low dimensionality (quasi-one-dimensional electron systems, for instance) as molecular and biological polymers, which can also be excited by chemical interaction with the surrounding medium, may display the morphological transition and the chaotic-like behavior we have described. The application of NESOM to the study of nonequilibrium electron system in quantum-wires have been reported in reference [10], where a derivation and a detailed analysis of the dielectric function as well as the instability of collective modes (plasmons) was carried out. In the case of biopolymers, the efficient mechanism in action resides in the so-called dark biological processes (chemical reactions involving enzyme catalysis, in particular, the hydrolysis of ATP). Therefore, since excited states in these systems can be generated *via* two different mechanisms (electromagnetic radiation and chemical reaction), lower radiation levels could produce the instability without crippling material damage. This is a quite interesting result that goes in the direction of the ideas advanced by Szent-György, that mobile electrons have an enormous relevance in biosystems, playing a fundamental role in the working of life, and that the resulting carrier conductivity in extended proteins molecules may result in the building of higher structures [26] (as the situation we have described in this paper).

We have shown on the basis of the analysis of the static wavenumber-dependent dielectric function, that with increasing power of the pumping source beyond the critical point (first bifurcation), more and more subharmonics of the fundamental mode contribute to the construction of the static charge density wave. As the pumping power increases the number of these components grows in such a way that the system could display chaotic behavior, phenomenon that may be called turbulent-like chaos, in analogy with the old Landau's theory of turbulence, who discussed the possible emergence of turbulence in terms of an excess of modes emerging in the fluid.

One of us (SAH) is grateful to Prof. Nelson Duran of the Institute of Chemistry at Unicamp, for helpful discussions on the subject of reference [25]. We acknowledge financial support provided to our Group in different opportunities by the São Paulo State Research Foundation (FAPESP), the National Research Council (CNPq), Unicamp Foundation (FAEP), the Ministry of Planning (Finep), IBM-Brasil, and the John Simon Guggenheim Memorial Foundation.

Appendix A: Characterization of bifurcation points

Determination of critical points of instability in nonlinear systems is, in general, a difficult task. The mathematical approach is based, as known, on the use of Lyapounov's theory. In nonlinear nonequilibrium thermodynamics Glansdorff-Prigogine (in)stability criterion, based on the change of sign of the quantity called excess entropy production function [27], is available. We recall that because of Prigogine's theorem of minimum entopy production in the strictly linear (Onsagerian) regime, the disorganized state (thermal chaos) is always stable. In the nonlinear regime its instability can follow when the system is driven by external exciting sources and Onsager's symmetry laws are no longer valid (these results can be generalized in the scope of Informational Statistical Thermodynamics, as shown in the third of Ref. [14]).

In the case we have considered in the main text, we can use an alternative approach based on response function theory, in close analogy with the case of phase transitions in equilibrium. In the latter case the critical (or transition) point is characterized by a singularity in a particular physical property as, for example, an infinite value of the specific heat in transitions involving structural changes, an infinite value of the magnetic susceptibility in a ferromagnetic transition, etc. Moreover, characteristics of the transition can also be derived: in the first case just mentioned, since $\Delta Q = C\Delta T$, by definition of the specific heat C, where ΔQ is the heat provided and ΔT the change in temperature, at the transition point as C goes to infinity $\Delta T = 0$ and the temperature remains constant as the transition proceeds; in the second case, the magnetic energy being $\mathbf{H} \cdot \mathbf{B}/2 = \mu_0 |\mathbf{H}|^2 (1 + \chi_M)/2$, at the critical point the magnetic susceptibility χ_M goes to infinity and then $\mathbf{H} = 0$, implying in that a spontaneous ferromagnetic magnetization $M \neq 0$ must emerge.

Consider now the case of nonequilibrium systems arbitrarily away from equilibrium, as the one we have considered in the main text. In this case we can introduce ideas having a close analogy with the case of phase transitions, however keeping in mind that the role of phases in equilibrium is now played by stationary dissipative structures. For the carriers' system described in this paper, since $\epsilon(\mathbf{Q}, 0)$ is the static wavenumber-dependent dielectric function and the electric energy is given by

$\mathbf{E}(\mathbf{Q}) \cdot \mathbf{D}(\mathbf{Q})/2 = \epsilon^{-1}(\mathbf{Q}, 0)|\mathbf{D}(\mathbf{Q})|^2/2$, at the point where ϵ^{-1} goes to infinity, that is when ϵ goes to zero, $\mathbf{D}$ must also go to zero, implying in the emergence of a spontaneous and space-dependent electric polarization $\mathbf{P} \neq 0$, namely, the charge density wave we have evidenced. In this way, this is completely analogous to the case of electrical polarizable phase transitions in equilibrium, with $\mathbf{Q} = 0$ for ferroelectrics; $|\mathbf{Q}| = \pi/a$ for antiferroelectrics (a is the lattice parameter in a given direction) and arbitrary $\mathbf{Q}$ for helical-electric materials.

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