



STEADY-STATE POLARIZATION WAVES IN PHOTOEXCITED n-DOPED SEMICONDUCTORS

Antonio Sergio Esperidião  
Departamento de Física, Universidade Federal da Bahia,  
40021 - Salvador, BA, Brasil

and

Aurea R. Vasconcellos and Roberto Luzzi  
Instituto de Física, Universidade Estadual de Campinas,  
C.P. 6165; 13081 - Campinas, SP, Brasil

(Received Feb. 1, 1990 by C.E.T. Gonçalves da Silva)

We show that, in n-doped semiconductors under continuous laser illumination the homogeneous steady-state of carriers becomes, in the degenerate regime, unstable against the formation of spatial order at the macroscopic level. Beyond a critical distance from equilibrium this nonequilibrium phase transition occurs, corresponding to the formation of a polarization wave of electrons and holes. It is the result of the interplay of collective and nonlinear dissipative effects in the far-from-equilibrium system.

Nonlinear effects in open systems are nowadays a subject of large physical and technical interest. Nonlinearity is the source of new and unexpected behavior in matter.<sup>(1,2)</sup> Decades ago, Turing showed that morphological transitions may develop as a result of diffusion instability in open systems governed by nonlinear kinetic laws.<sup>(3)</sup> We consider here a question of this type in the case of photoexcited semiconductors, aiming to put into evidence the occurrence of a spatially self-organized structure of the carrier density. For that purpose we resort to the nonequilibrium statistical operator method<sup>(5)</sup> (NSOM)<sup>(4)</sup> in Zubarev's approach<sup>(5)</sup> which yields a nonlinear quantum transport theory, that can be considered a far-reaching generalization of the Hilbert-Chapman-Enskog's and Mori's methods.<sup>(4)</sup> It provides the nonlinear equations of evolution of the macroscopic state of the system, and then a way to evidence and describe at a microscopic level nonlinear instabilities. We have applied Zubarev's NSOM to the study of the evolution of a polar semiconductor under continuous laser light illumination,<sup>(6)</sup> where a brief review of the method is given.

Consider a n-doped polar semiconductor with an electron concentration  $n_0$  high enough for them to behave as itinerant carriers in the conduction band. Under the action of C.W. laser light, a concentration  $n$  of photoinjected electrons and holes is produced in pairs by photon absorption.

As shown in Ref. 6 after a sufficient delay time a homogeneous steady state follows, characterized by a stationary concentration  $n$  and quasi-temperature  $\beta^{-1}$ . Assuming a good thermal contact of the system with the reservoir at temperature  $T_0$ , it follows that  $\beta^{-1} \approx kT_0$ . The equations for  $n$  and  $\beta^{-1}$  of Ref. 6 remain valid for the extrinsic semiconductor, except that it must be kept in mind that the concentration of electrons is now  $n_0 + n$ . To test the stability of the homogeneous steady state against the formation of spatial ordering, we consider an infinitesimal spatial fluctuation imposed on the system. For that purpose we enlarge the basic set of NSOM-variables with the inclusion of the nondiagonal elements of the one-quasiparticle density matrix, namely

$$n_{\vec{k}\vec{a}}^e(t) = \text{Tr} \left\{ C_{\vec{k}\vec{a}}^\dagger C_{\vec{k}} \rho_\epsilon(t) \right\}, \quad (1a)$$

$$n_{\vec{k}\vec{a}}^h(t) = \text{Tr} \left\{ h_{-\vec{k}\vec{a}}^\dagger h_{-\vec{k}} \rho_\epsilon(t) \right\}, \quad (1b)$$

where  $C(C^\dagger)$  and  $h(h^\dagger)$  are annihilation (creation) operators for electrons and holes in plane-wave states and  $\rho_\epsilon$  is Zubarev's NSO.

The NSOM-equations of evolution for the quantities of Eqs. (1), in Zubarev's NSOM linear theory of relaxation and using the random phase

approximation (RPA) to deal with Coulomb interaction, are

$$i\hbar \frac{\partial}{\partial t} n_{\vec{k}\vec{a}}^e(t) = \Delta E_{\vec{k}\vec{a}}^e n_{\vec{k}\vec{a}}^e + 2V(Q)\Delta f_{\vec{k}\vec{a}}^e n(Q) + iB_{\vec{k}\vec{a}}^{e,h} n_{\vec{k}\vec{a}}^h - iB_{\vec{k}\vec{a}}^{e,h} n_{\vec{k}\vec{a}}^e + N_{\vec{k}\vec{a}}^e \quad (2a)$$

and a similar one for holes (2b)

In Eqs. (2),  $\Delta E_{\vec{k}\vec{a}}^{e(h)} = \epsilon_{\vec{k}\vec{a}}^{e(h)} - \epsilon_{\vec{k}\vec{a}}^{e(h)}$ ;  
 $\epsilon_{\vec{k}}^{e(h)} = \hbar^2 k^2 / 2m_{e(h)}$ ;  $V(Q) = 4\pi e^2 / \epsilon_0 V Q^2$ ,

where  $\epsilon_0$  is the static (background) dielectric constant and  $V$  the volume of the system;  $\Delta f_{\vec{k}\vec{a}}^{e(h)} = f_{\vec{k}\vec{a}}^{e(h)} - f_{\vec{k}\vec{a}}^{e(h)}$ , with  $f_{\vec{k}}^{e(h)}$  being Fermi-Dirac distribution functions. Further,

$$n(Q) = \sum_{\vec{k}} \left[ n_{\vec{k}\vec{a}}^e + n_{\vec{k}\vec{a}}^h \right] \quad (3)$$

is, in units of the electron charge, the Q-wavenumber component of the charge density.  $N_{\vec{k}\vec{a}}^{e(h)}$  are terms bilinear in the

quantities of Eqs. (1), and coefficients  $B$  contain the effect of laser light absorption, radiation recombination, and carrier-phonon interactions. The latter produces relaxation effects much smaller than the others and is neglected. Then  $B_{\vec{k}\vec{a}}^{e,h} = B_{\vec{k}\vec{a}}^{h,h} = B_{\vec{k}\vec{a}}^{e,e}$  and  $B_{\vec{k}\vec{a}}^{h,e} = B_{\vec{k}\vec{a}}^{e,h}$ , with

$$B_{\vec{k}\vec{a}}^{e(h)} = A_L \delta(\epsilon_{\vec{k}}^x + E_g - \hbar\omega_L) + A_R (\epsilon_{\vec{k}}^x + E_g) f_{\vec{k}}^{e(h)}, \quad (4)$$

where  $A_L$  and  $A_R$  depend on the matrix elements of the interaction potential of carriers with the laser and recombination fields respectively,  $\epsilon_{\vec{k}}^x = \hbar^2 k^2 (m_e^{-1} + m_h^{-1}) / 2$ , and  $E_g$  is the energy gap.

The steady-state solution (fix point) of Eqs. (2) corresponds to

$n_{\vec{k}\vec{a}}^{e(h)} = 0$  (or  $n(Q) = 0$ ), i.e. the homogeneous state. Hence, the linearized equations around the steady-state are in linear stability analysis<sup>(1,7)</sup> Eqs. (2) putting  $N_{\vec{k}\vec{a}}^{e(h)} = 0$ . We explicitly look

for an instability against a static inhomogeneity, and thus we need to analyse the existence of a null eigenvalue of the linearized set of Eqs. (2).<sup>(7)</sup> Lengthy and elaborate Algebra leads us to the secular equation

$$n(Q)\epsilon(Q) = 0, \quad (5)$$

where

$$\epsilon(Q)/\epsilon_0 = 1 - V(Q) \sum_{\vec{k}} M(\vec{k}, Q) D^{-1}(\vec{k}, Q), \quad (6)$$

with

$$M(\vec{k}, Q) = \left[ \Delta f_{\vec{k}\vec{a}}^e \Delta E_{\vec{k}\vec{a}}^h + \Delta f_{\vec{k}\vec{a}}^h \Delta E_{\vec{k}\vec{a}}^e \right] - i \left[ \Delta f_{\vec{k}\vec{a}}^e - \Delta f_{\vec{k}\vec{a}}^h \right] \left[ B_{\vec{k}\vec{a}}^e + B_{\vec{k}\vec{a}}^h \right], \quad (7a)$$

$$D(\vec{k}, Q) = \left[ \Delta E_{\vec{k}\vec{a}}^e + iB_{\vec{k}\vec{a}}^h \right] \left[ \Delta E_{\vec{k}\vec{a}}^h + iB_{\vec{k}\vec{a}}^e \right] - B_{\vec{k}\vec{a}}^e B_{\vec{k}\vec{a}}^h. \quad (7b)$$

It should be noted that  $\epsilon(Q)$  is the static dielectric function of the nonequilibrium stationary carrier system.

One solution of the secular equation, Eq. (5), is  $n(Q) = 0$ , i.e. the one corresponding to the uniform state, and another with  $n(Q) \neq 0$  (spatial ordering in the form of a nonequilibrium stationary charge density wave) is possible if  $\epsilon(Q) = 0$ . Its imaginary part is identically null, and then we must look for a zero of its real part, i.e.

$$\text{Re } \epsilon(Q) = 1 - V(Q) \sum_{\vec{k}} \nu(\vec{k}, Q) \Delta^{-1}(\vec{k}, Q) = 0, \quad (8)$$

where

$$\nu(\vec{k}, Q) = \left[ \Delta f_{\vec{k}\vec{a}}^e \Delta E_{\vec{k}\vec{a}}^h + \Delta f_{\vec{k}\vec{a}}^h \Delta E_{\vec{k}\vec{a}}^e \right] \Delta E_{\vec{k}\vec{a}}^e \Delta E_{\vec{k}\vec{a}}^h + \left[ \Delta f_{\vec{k}\vec{a}}^h - \Delta f_{\vec{k}\vec{a}}^e \right] \left[ B_{\vec{k}\vec{a}}^e + B_{\vec{k}\vec{a}}^h \right] \left[ \Delta E_{\vec{k}\vec{a}}^h - \Delta E_{\vec{k}\vec{a}}^e \right], \quad (9a)$$

$$\Delta(\vec{k}, Q) = \left[ \Delta E_{\vec{k}\vec{a}}^e \cdot \Delta E_{\vec{k}\vec{a}}^e \right]^2 + \left[ B_{\vec{k}\vec{a}}^h \Delta E_{\vec{k}\vec{a}}^h - B_{\vec{k}\vec{a}}^e \Delta E_{\vec{k}\vec{a}}^e \right]^2. \quad (9b)$$

Numerical calculations are performed using parameters characteristic of GaAs, and a pumping laser with photon energy of 2.4 eV and variable laser power. Our results show that there exists a critical laser power, or, in other words, a critical concentration  $n_c$ , for each value of  $n_0$  that satisfies Eq. (8) and therefore characterizes the instability of the homogeneous steady state. Figure 1 shows the value of the critical concentration, for  $n_0 = 10^{17} \text{ cm}^{-3}$  and  $5 \times 10^{17} \text{ cm}^{-3}$ , for several values of the wavenumber  $Q$ . The calculation was done taking the carriers in the very degenerate regime such that their distribution functions can be approximated by step functions centered at the quasi-chemical potentials. For  $Q = 0$ , the critical carrier photoconcentration is given by

$$\left[ \left[ \frac{2m_h}{m_h - m_e} \right]^3 - 1 \right] n_{\text{crit}} = n_0 \quad (10)$$

It should be noted that there is no solution to Eq. (8) in the case of an intrinsic semiconductor. In this case

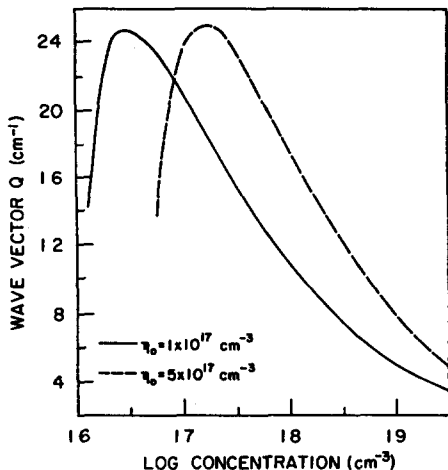


Fig. 1: The critical value of the photoinjected concentration for the different values of the wavenumber of the emerging polarization wave, and two values of  $n_0$ . The critical concentration of photoinjected carriers is  $9 \times 10^{15} \text{ cm}^{-3}$  in the first case and  $4.5 \times 10^{15} \text{ cm}^{-3}$  in the second.

what is obtained is a metallic to non-metallic transition in the extremely degenerate regime.<sup>(8)</sup> There is no instability in the case of a p-doped material, and also in the n-doped semiconductor at high temperatures (classical limit). This tells us that an excess of light particles is necessary to ensure the formation of the spatial structure but such ordering is destroyed by thermal agitation, i.e. formed in the degenerate regime  $n(Q)$  tends to zero with increasing carrier quasitemperature. The first instability follows for  $Q$  going to zero (infinite wavelength) at a minimum critical laser power  $I_L$ , and for subsequent values of  $Q$  for increasing  $I_L$  above  $I_L^*$ . It is worth noticing that this characteristic implies that the carrier system goes along a nonequilibrium thermodynamic path from thermal chaos (at low  $I_L$ ), to order (spatial structure at  $I_L^*$ ), to what has been termed turbulent nonequilibrium chaos (at higher  $I_L$ ), i.e. abundance of macroscopic length scales so that the system appears chaotic. For the realistic case of a finite system boundary conditions must be imposed;  $Q$  should admit the values  $k\pi/L$ ,  $k = 1, 2, \dots$ , if  $L$  is the dimension of the sample in the direction of  $Q$ . Furthermore, ours is an isotropic model and thus when crystalline symmetry is considered it would introduce additional restrictive conditions and, conjecturably, lead to critical values

of  $Q$  corresponding to points of high symmetry in the Brillouin zone. Since it should be expected that the polarization wave in the carrier system would drive along the ions, a long range order in the whole material would follow. The situation may then be considered akin to the nonthermal recrystallization of ion implanted semiconductors proposed by van Vechten.<sup>(9)</sup>

We have also calculated the electronic Raman spectrum to be expected from this system, depicted in Figure 2. As we have previously shown<sup>(10)</sup> it consists of four bands due to scattering by: (1) optical plasmons, (2) quasi-particle excitations, (3) upper acoustic plasmons, and (4) lower acoustic plasmons the last two being shown in Fig. 2. It is observed in this case of an n-doped semiconductor, that, on approaching the critical point, band (4) merges and disappears into band (3). Since the former is associated to the collective motion of holes interacting through the screened part of Coulomb

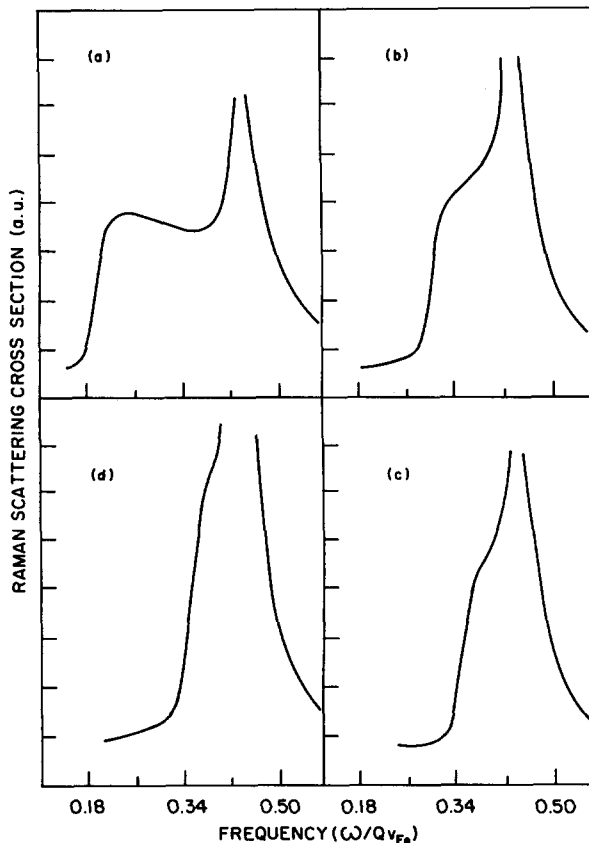


Fig. 2: The Raman scattering cross section for the case  $n_0 = 10^{17} \text{ cm}^{-3}$ . Clockwise from top left: (a)  $n = 10^{16} \text{ cm}^{-3}$ ; (b)  $n = 5 \times 10^{16} \text{ cm}^{-3}$ ; (c)  $n = 8 \times 10^{16} \text{ cm}^{-3}$ , and (d)  $n = 10^{17} \text{ cm}^{-3}$ .  $Q$  is  $0.5 \text{ cm}^{-1}$ , and the critical concentration is  $n_{\text{crit}} \approx 9 \times 10^{15} \text{ cm}^{-3}$ .

interaction and the latter to that of electrons,<sup>(10)</sup> this result suggests that at the transition there is formation of charge densities of electrons and of holes clamped together, in what may be termed a polarization wave.

ACKNOWLEDGEMENTS - One of us (ASE) is a Brasil Ministry of Education (CAPES) pre-doctoral fellow; the other two (ARV,RL) are Brasil National Research Council (CNPq) fellows.

## REFERENCES

- 1 G. Nicolis and I. Prigogine, *Self-Organization in Nonequilibrium Systems* (Wiley, New York, 1977).
- 2 H. Haken, *Synergetics* (Springer, Berlin-Heidelberg, 1978).
- 3 A.M. Turing, *Phil. Trans. Roy. Soc. B237*, 37 (1952).
- 4 For a review of the NSOM and its applications see: R. Luzzi and A.R. Vasconcellos, *Fortsch. Phys.* No. 1, Band 39 (in press); also *Rev. Brasil. Fis.* 15, 106 (1985); *ibid* 16, 495 (1986); *ibid* 17, 601 (1987).
- 5 D.N. Zubarev, *Neravnovesnaia Statisticheskaiia Termodinamika* (Izd. Nauka, Moskva, 1971) [*Nonequilibrium Statistical Thermodynamics* (Plenum, New York, 1974)].
- 6 T. Tomé, A.R. Vasconcellos and R. Luzzi, *Physica* 144B, 376 (1987).
- 7 P. Davies, *Nonlinear Integral and Differential Equations* (Dover, New York, 1962); also in Ref. (1), Part II, Section 6.
- 8 A.S. Esperidião, A.R. Vasconcellos and R. Luzzi, *Solid State Commun.* (in press).
- 9 J.A. van Vechten, *J. Phys. (Paris)* C41, 4 (1980); *Solid State Commun.* 39, 1835 (1981).
- 10 A.S. Esperidião, A.R. Vasconcellos and R. Luzzi, *Solid State Commun.* (in press).