Solid State Communications, Vol. 73, No. 10, pp. 673-676, 1990.
Printed in Great Britain.

0038-1098/90$3.00+.00
Pergamon Press plc

# NONEQUILIBRIUM PHASE TRANSITION IN PHOTOINJECTED PLASMA IN SEMICONDUCTORS

Antonio Sergio Esperidião
Instituto de Física, Universidade Federal da Bahia
40211 - Salvador, BA, Brasil

and

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

(Received 31 August 1989 by Cylon E. T. Goncalves da Silva)

We analize the stability of the nonlinear nonequilibrium thermodynamic steady state of a double component plasma generated by continuous laser light illumination in direct-gap polar semiconductors. It is shown that in the extremely degenerate regime there occurs a transition to a non-metallic nonequilibrium dissipative structure. In the intermediate degenerate regime there is a coexistence of the non-metallic nonequilibrium phase with a metallic one. Above a certain temperature, for each concentration, the carrier system is almost purely metallic. The non-conducting phase is produced as a result of collective plus nonlinear dissipative effects in the far-from-equilibrium system.

Formation in semiconductors of a uniform steady-state double-component plasma of electrons and holes (carriers) in the lattice background is possible through continuous laser illumination. The laser intensity should be high enough to produce a concentration of photoinjected carriers such that the system is on the metallic side of Mott transition (typically $10^{16}$ $cm^{-3}$ and higher). This nonequilibrium open system has quite interesting characteristics, like the presence in its excitation spectrum of two branches of acoustic plasmons.[1] Further, we recall that open systems in sufficiently far-from-equilibrium conditions, where they are governed by nonlinear kinetic laws, may become unstable against the formation of order on the macroscopic level.[2,3] The system we are considering belongs to this class and thus it requires stability analysis. We test the uniform solution against the formation of a nonhomogeneous structure. We resort to normal mode analysis, i.e. the study of the eigenvalue spectrum of the linearized equations of motion resulting after the imposition of a small deviation on the steady state.[4] Since we are looking for a new stationary solution we must try for the existence of a null eigenvalue of the linearized equations, i.e. zero oscillation frequency and infinite lifetime. The equations of evolution for the variables that describe the macroscopic state of the system are obtain from the nonlinear quantum transport theory that is derived from the nonequilibrium statistical operator method (NSOM)[5], and Zubarev's approach is used.[6] (see also Ref. 7)

Consider an intrinsic polar semiconductor illuminated by continuous laser light. After a transient time there follows a uniform steady state characterized by a quasitemperature $\beta^{-1}$[7] and a concentration n of carriers. Next we add to the basic set of NSOM-variables the set of those composed by the nondiagonal elements of the single quasi-particle density matrix, namely

$$n^{e}_{\vec{k}\vec{Q}}(t) = Tr\langle C^{\dagger}_{\vec{k}+\vec{Q}} C_{\vec{k}} \rho_{\varepsilon}(t)\rangle \qquad (1a)$$

$$n^{h}_{\vec{k}\vec{Q}} = Tr\langle h_{-\vec{k}-\vec{Q}} h^{\dagger}_{-\vec{k}} \rho_{\varepsilon}(t)\rangle \quad , \qquad (1b)$$

where $C(C^{\dagger})$ and $h(h^{\dagger})$ are anihilation (creation) operators of electrons and holes in plane-wave states, and $\rho_{\varepsilon}(t)$ is Zubarev's NSO. The NSOM-equations of evolution for these quantities, linearized around the homogeneous steady state $n^{e(h)}_{kQ} = 0$, are

$$i\hbar \frac{\partial}{\partial t} n^{e}_{\vec{k}\vec{q}}(t) = \Delta E^{e}_{\vec{k}\vec{q}}\, n^{e}_{\vec{k}\vec{q}} - 2V(\vec{Q})\,\Delta f^{e}_{\vec{k}\vec{q}}\, n(\vec{Q}) + $$

$$+ i B^{eh}_{\vec{k}\vec{q}}\, n^{h}_{\vec{k}\vec{q}} - i B^{ee}_{\vec{k}\vec{q}}\, n^{e}_{\vec{k}\vec{q}} \quad , \qquad (2a)$$

$$i\hbar \frac{\partial}{\partial t} n^{h}_{\vec{k}\vec{q}}(t) = -\Delta E^{h}_{\vec{k}\vec{q}}\, n^{h}_{\vec{k}\vec{q}} + 2V(\vec{Q})\,\Delta f^{h}_{\vec{k}\vec{q}}\, n(\vec{Q}) + $$

$$+ i B^{he}_{\vec{k}\vec{q}}\, n^{e}_{\vec{k}\vec{q}} - i B^{hh}_{\vec{k}\vec{q}}\, n^{h}_{\vec{k}\vec{q}} \quad , \qquad (2b)$$

where $\Delta E^{e(h)}_{\vec{k}\vec{q}} = \varepsilon^{e(h)}_{\vec{k}+\vec{q}} - \varepsilon^{e(h)}_{\vec{k}}$;

$\varepsilon^{h}_{\vec{k}} = \hbar^2k^2/2m_h$, $\varepsilon^{e}_{\vec{k}} = E_g + \hbar^2k^2/2m_e$,

$V(Q) = 4\pi e^2/\varepsilon_o V Q^2$, with $\varepsilon_o$ the background static dielectric constant and V the volume of the system; $E_g$ is the energy gap:

$$n(\vec{Q}) = \sum_{\vec{k}} (n^{e}_{\vec{k}\vec{q}} + n^{h}_{\vec{k}\vec{q}}) \qquad (3)$$

is, in units of the electron charge, the Q-wavenumber component of the carrier charge density; $\Delta f^{e(h)}_{\vec{k}\vec{q}} = f^{e(h)}_{\vec{k}+\vec{q}} - f^{e(h)}_{\vec{k}}$, $f^{e(h)}_{\vec{k}}$ beingFermi-Dirac distribution functions, since, as noted, the carriers are in internal equilibrium. Further, coefficients B contain dissipative effects associated to laser light absorption, radiative recombination, and carrier-phonon interaction. The latter produces relaxation effects much smaller than those of recombination and is neglected.[7] Then, $B^{eh} = B^{hh} = B^{e}$ and $B^{he} = B^{ee} = B^{h}$ are given by

$$B^{e(h)}_{\vec{k}\vec{q}} = A_L\, \delta(\varepsilon^{x}_{\vec{k}} + E_g - \hbar\omega_L) + A_R(\varepsilon^{x}_{\vec{k}} + E_g) f^{e(h)}_{\vec{k}} , \qquad (4)$$

where $A_L$ and $A_R$ are coefficients depending on the matrix elements of the interaction of the carriers with the laser and recombination radiation fields, and $\varepsilon^{x}_{k} = \hbar^2k^2/2m_x$, with $m_x^{-1} = m_e^{-1} + m_h^{-1}$. Coulomb interaction has been treated in the Random Phase Approximation (RPA).

Putting both equations (2) equal to zero, the case of null eigenvalue, we solve them and adding both quantities $n^{e(h)}_{\vec{k}\vec{q}}$ and summing over $\vec{k}$, we find the secular equation

$$n(\vec{Q})\,\varepsilon(\vec{Q}) = 0 \quad , \qquad (5)$$

where

$$\text{Real}\ \varepsilon(\vec{Q})/\varepsilon_o = 1 - V(\vec{Q}) \sum_{\vec{k}} \nu(\vec{k},\vec{Q})\,\Delta^{-1}(\vec{k},\vec{Q}) \quad , \qquad (6)$$

with

$$\nu(\vec{k},\vec{Q}) = (\Delta f^{e}_{\vec{k}\vec{q}}\,\Delta E^{h}_{\vec{k}\vec{q}} + \Delta f^{h}_{\vec{k}\vec{q}}\,\Delta E^{e}_{\vec{k}\vec{q}})\,\Delta E^{e}_{\vec{k}\vec{q}}\,\Delta E^{h}_{\vec{k}\vec{q}} +$$

$$+ (\Delta f^{h}_{\vec{k}\vec{q}} - \Delta f^{e}_{\vec{k}\vec{q}})(B^{e}_{\vec{k}\vec{q}} + B^{h}_{\vec{k}\vec{q}})(\Delta E^{h}_{\vec{k}\vec{q}}\, B^{h}_{\vec{k}\vec{q}} - \Delta E^{e}_{\vec{k}\vec{q}}\, B^{e}_{\vec{k}\vec{q}}) \quad , \qquad (7a)$$

$$\Delta(\vec{k},\vec{Q}) = (\Delta E^{e}_{\vec{k}\vec{q}}\,.\,\Delta E^{h}_{\vec{k}\vec{q}})^2 + (B^{h}_{\vec{k}\vec{q}}\,\Delta E^{h}_{\vec{k}\vec{q}} - B^{e}_{\vec{k}\vec{q}}\,\Delta E^{e}_{\vec{k}\vec{q}})^2 \qquad (7b)$$

and Im $\varepsilon(\vec{Q}) = 0$. Here $\varepsilon(\vec{Q})$ is the static and wave-vector dependent dielectric function of the system, obtained in RPA and including relaxation effects contained in coefficients B that couple electrons and holes. This expression goes over Lindhart dielectric function for a system of two types of free carriers when putting B = 0.[8]

Eq. (5) tells us that one solution is $n(\vec{Q}) = 0$, i.e. the one corresponding to the homogeneous state. A solution $n(\vec{Q}) \neq 0$, i.e. corresponding to formation of spatial ordering, follows for $\varepsilon(\vec{Q}) = 0$. Lengthy but straightforward calculations shows that in the classical limit and small Q,

$$\varepsilon(\vec{Q})/\varepsilon_o \simeq 1 + Q^2_{DH}(\beta,n)/Q^2 \quad , \qquad (8)$$

where $Q_{DH}$ is Debye-Hückel screening wavenumber, $Q^2_{DH} = 8\pi e^2 n\beta/m_x\varepsilon_o$, and thus Eq. (8) has no zero. On the extremely degenerate regime (very low temperatures and high concentration) and small Q we find

$$\varepsilon(\vec{Q})/\varepsilon_o \simeq \varepsilon_p(\beta,n) + Q^2_o(\beta,n)\,Q^2_o(\beta,n)/Q^2, \qquad (9)$$

where

$$\varepsilon_p(\beta,n) = 1 - \frac{4\pi e^2\hbar^4}{\varepsilon_o V} \sum_{\vec{k}} k^2 \left[ \frac{f'_{\vec{k}e} + f'_{\vec{k}h}}{(m_e B^{h}_{\vec{k}} - m_h B^{e}_{\vec{k}})^2} + \right.$$

$$\left. + \frac{(m_e f'_{\vec{k}h} - m_h f'_{\vec{k}e})(B^{e}_{\vec{k}} + B^{h}_{\vec{k}})}{(m_e B^{h}_{\vec{k}} - m_h B^{e}_{\vec{k}})^3} \right] \quad , \qquad (10a)$$

and

$$Q_o^2(\beta,n) = \frac{4\pi e^2\hbar^4}{\varepsilon_o V}\sum_{\vec{k}}\left(m_h f'_{\vec{k}e} - m_e f'_{\vec{k}h}\right)\frac{B^e_{\vec{k}}+B^h_{\vec{k}}}{m^e B^h_{\vec{k}} - m_h B^e_{\vec{k}}}\,, \qquad (10b)$$

with

$$f^{e}_{\vec{k}e(h)} = \partial f^{e(h)}_{\vec{k}}/\partial\varepsilon^{e(h)}_{\vec{k}}\,.$$

Hence, $\varepsilon_p$ and $Q_o$ are finite positive numbers and consequently there is no zero of $\varepsilon(\vec{Q})$ in this limit and so is the case in any intermediate regime. Consequently the photoexcited intrinsic semiconductor shows a uniform electronic steady state stable against any nonuniform spatial fluctuation.

However, we are in conditions to put into evidence another kind of transition between homogeneous steady states. According to Eq. (10b) $Q_o$ is dependent on the nonequilibrium state of the system and in the extremely degenerate regime, when f' becomes a delta function centered on the Fermi momentum, $Q_o$ is null. Therefore, the static dielectric function, for Q going to zero, is finite (equal to $\varepsilon_p$) and the carrier system is no longer metallic, the metallic property characterized by an infinite dielectric constant at Q = 0.

On leaving the extremely degenerate regime, $Q_o$ becomes nonnull. In Fig. 1 we show the Q-dependent dielectric function of Eq. (6) (full line) for carrier quasitemperatures $T^*$ of 2K and 10K. For comparison we have drawn (dashed line) Lindhart dielectric function for the double free carrier system, i.e. B = 0 in Eq. (6). We have used parameters characteristic of GaAs and $n = 10^{16}$ cm$^{-3}$, obtained by illumination with laser light of photon energy $\hbar\Omega_L$ = 2.4 eV and power intensity $I_L \simeq 4$ Watt/cm$^2$.

For $Q_o = 0$ the Coulomb potential is not screened and then electrons and holes interact through the bare Coulomb interaction. The resulting nonmetallic carrier system should resemble a nonequilibrium phase with electrons and holes bounded in a nonconducting polarizable state. The screening length becomes finite in the degenerate regime and tends to the Debye-Hückel value in the classical limit [Cf. Eq. (8)], when the carriers would then be in a near complete metallic state. Inspection of the curves in Fig. 1 tells us that in fact with increasing carrier quasi-temperature $\varepsilon(Q)$ tends to coincide with Lindhart's $\varepsilon_L(Q)$. This suggests that between the nonmetallic state at the extremely degenerate regime and the classical regime the carrier system is in a nonequilibrium thermodynamic phase with coexistence of carriers in both metallic and nonmetallic states. Using a simple model of two independent fluids for both states, we can obtain an estimative of the fraction of carriers in the condensate (the polarizable state of bounded electrons and holes); $\Delta = n^*/n$ ($n^*$ being the density of carriers in the condensate) given by

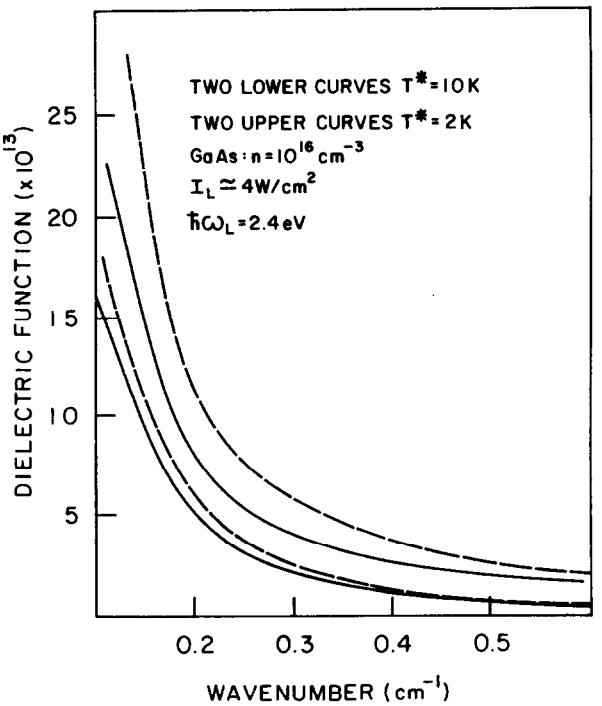


Fig. 1: The dielectric function of Eq. (6) (full lines) and Lindhart's dielectric function for the two component free carrier system (dashed lines).

$$\Delta(\beta,n) = \lim_{Q\to 0}\frac{\varepsilon_L(Q)-\varepsilon(Q)}{\varepsilon_L(Q)}\,. \qquad (11)$$

We show this value (obtained for Q = 1 cm$^{-1}$) as a function of the carrier quasitemperature, for $n = 10^{16}$ cm$^{-3}$, in Fig. 2. The resulting curve can be approximated by the expression $\Delta \simeq \exp[-T^*/T_c(n)]$, where we find $T_c \simeq 3.6$ K. Thus, for values of $T^*$ of

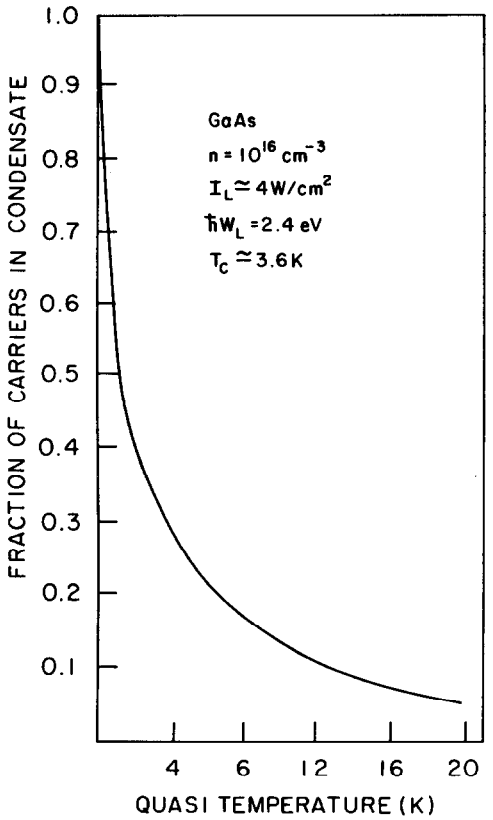


Fig. 2: Fraction of carriers in the nonmetallic polarizable state as a function of carrier quasitemperature.

the order and larger than this $T_c$ the system is in an almost completely metallic state.

In the isotropic model we used the polarizable (nonmetallic) structure seems to correspond to two interpenetrated infinite wavelength Wigner-like lattices clamped together. It is conjecturable that introducing, say, a tight binding approximation for the hole states the condensed dissipative structure would resemble a fluid of Frenkel excitons. In summary, at a given carrier quasitemperature with increasing laser power, and thus increasing concentration of photoinjected carriers, as known excitons are formed at low concentration, followed by their ionization on approaching the point of occurrence of Mott transition. Next, according to our results, the carriers fluid partially condense in a polarizable nonconducting state and partially remains in an itinerant conducting state. Except in the highly degenerate regime the carriers are in large proportion forming the metallic fluid. [Cf. Fig. 2]

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. A.S. Esperidião, A.R. Vasconcellos, and R. Luzzi, Solid State Commun. (in press).
2. G. Nicolis and I. Prigogine, *Self-Organization in Nonequilibrium Systems* (Wiley, New York, 1977).
3. H. Haken, *Synergetics* (Springer, Berlin - Heildelberg, 1978).
4. E.g. in Ref. (2), Part II, Section 6; also H.T. Davies, *Nonlinear Differential and Integral Equations* (Dover, New York, 1962).
5. For a review of the NSOM and its applications see: R. Luzzi and A.R. Vasconcellos, Fortsch. Phys. (submitted); also Rev. Brasil. Fis. **15**, 106 (1985); ibid **16**, 495 (1986); ibid **17**, 601 (1987).
6. D.N. Zubarev, *Neravnovesnaia Statisticheskaia Thermodinamica* (Izd. Nauka, Moskwa, 1971) [*Nonequilibrium Statistical Thermodynamics* (Plenum, New York, 1974)].
7. T. Tomé, A.R. Vasconcellos, and R. Luzzi, Physica **144B**, 376 (1987).
8. E.g. C. Kittel, *Quantum Theory of Solids* (Wiley, New York, 1963).