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ACOUSTIC-PLASMON BRANCHES IN PHOTOEXCITED SEMICONDUCTORS

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We analyze the spectra of electronic elementary excitations in a double component plasma of photoinjected electrons and holes in semiconductors under continuous laser illumination. We show that in the resulting nonequilibrium steady state four types of elementary excitations of the photoinjected carrier system can be identified. Two are the expected single quasi-particle excitations and the higher frequency plasma wave, and two novel ones at low frequencies labelled as acoustic plasma waves that are well defined under particular conditions. These acoustic branches are interpreted as corresponding to the collective motion of each type of carriers interacting through the screened part of Coulomb interaction.

The study of elementary excitations in solids has a long history, with a detailed study of the correlated electron motion - a classic in the literature on the subject - due to Pines and co-workers.[2] As part of their analysis, these authors have considered a two-component plasma (two types of different carriers in the positive background) and argumented on the possible existence of two branches of collective oscillations, labelled optical (the usual high energy) and acoustical (a new low energy) plasma waves. The first one is an oscillation out of phase of the charges with frequency $\Omega_{p\ell} = (\omega^2_{p\ell 1}+\omega^2_{p\ell 2})^{1/2}$ (dispersion neglected), where $\omega_{p\ell 1}$ and $\omega_{p\ell 2}$ are the plasma frequencies of each type of carrier. In the acoustic mode the particles would oscillate in phase with frequency $\omega_A = SQ$, where Q is the modulus of the wave-vector of the motion and S is the velocity $v_1[\omega_{p\ell}/\sqrt{3}\ \omega_{p\ell 1}]$ with v_1 being a suitable velocity of the light particles. This question was considered somewhat idealized in the sense of not being feasible in actual systems in equilibrium, and seemingly forgotten. However, such situation can be realized in nonequilibrium conditions in open semiconductors under intense c.w. laser illumination. The aim of this paper is to consider such situation, namely the elementary electron excitations in a highly photoexcited plasma in semiconductors. We show that there are three branches of collective elementary excitations, namely, the optical plasma wave with frequency $\Omega_{p\ell}$ and *two acoustic branches* to be identified with the collective movement of each of the light (electrons) and heavy (holes) particles interacting through a screened Coulomb interaction. These excitations are well defined and accompanied by a band of quasi-particle excitations for not too low values of wave-vector Q, merging and disappearing in the latter for Q going to zero.

We consider the model of a semiconductor with inverted parabolic bands illuminated with continuous laser light of frequency ω_L and power I_L, producing electron-hole pairs (carriers) on the metallic side of Mott transition (such metallic phase is attained when the carrier concentration is higher than a lower limit value, typically 10^{16} cm^{-3}).[2] To deal with this far-from-equilibrium system we resort to the nonequilibrium statistical operator method (NSOM) in Zubarev's approach.[3] We have applied this formalism to the study of ultrafast optical and transport properties of polar semiconductors, where a brief review of the method is given.[4,5] To the collective excitations of this system we calculate the Raman spectra resulting from scattering of the laser light by the carrier fluctuations. The differential Raman scattering cross section is proportional to the imaginary part of the inverse of the dynamic dielectric function $\varepsilon(\vec{Q},\omega)$. As known, scattering by the different elementary excitations gives rise to bands in the Raman spectrum. The calculation is done in the usual way using Maxwell equations and obtaining the polarization charge generated by a charge density probe of wave-vector $\vec{Q}$, frequency ω, and amplitude $-er_o$.[6] For that purpose we introduce in the basic set of NSOM-macrovariables

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

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

i.e. non-diagonal elements of the single-particle density matrices of electrons and of holes respectively; $\rho_{\varepsilon}(t)$ is Zubarev's NSO. The Fourier amplitude of the total charge density is (in units of the electron charge)

$$n(Q,t)=\sum_{\vec{k}} [n^{e}_{\vec{k}\vec{Q}}(t) + n^{h}_{\vec{k}\vec{Q}}(t)] \qquad (2)$$

Further, to complete the nonequilibrium thermodynamic description of the carriers' system we include the quasi-temperature $\beta^{-1}(t)$, and quasi-chemical potentials, $\mu_e(t)$ and $\mu_h(t)$.[4,7] They are obtained from the NSOM-transport equations[8] for the carriers' energy and density, given by Eqs. (15) and (27) in reference 7. Under continuous laser illumination after a transient period follows a steady state characterized by a constant quasi-temperature β^{-1}, which assuming a good thermal contact nearly equals that of the reservoir, and a constant photoinjected concentration of carriers, n, depending on I_L.[7]

Next we obtain the NSOM-equations of motion[8] for the macrovariables of Eq. (1), which are given by

$$\frac{\partial}{\partial t} n^{e}_{\vec{k}\vec{Q}}(t) = \frac{1}{i\hbar} r_o V(Q) [f^{e}_{\vec{k}+\vec{Q}} - f^{e}_{\vec{k}}] + \frac{1}{i\hbar}(\varepsilon^{e}_{\vec{k}+\vec{Q}} - \varepsilon^{e}_{\vec{k}})n^{e}_{\vec{k}\vec{Q}} + 2V(Q)(f^{e}_{\vec{k}+\vec{Q}} - f^{e}_{\vec{k}})n(\vec{Q})) + \frac{1}{\hbar} B^{e}_{\vec{k}\vec{Q}} n^{h}_{\vec{k}\vec{Q}} - \frac{1}{\hbar} B^{h}_{\vec{k}\vec{Q}} n^{e}_{\vec{k}\vec{Q}} \qquad (3)$$

and a similar one for holes, where $\varepsilon^{e(h)}_{\vec{k}} = \hbar^2k^2/2m_{e(h)}$; $f^{e(h)}_{\vec{k}}$ are Fermi-Dirac distribution functions with energy $\varepsilon^{e(h)}$, quasi-temperature β^{-1}, and quasi-chemical potentials $\mu_{e(h)}$; $V(Q) = 4\pi e^2/V\varepsilon_o Q^2$, where ε_o is the background static dielectric constant and V the volume of the system; the quantities $B^{e(h)}_{\vec{k}\vec{Q}}$ are

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

where A_L and A_R are connected with the matrix elements of the interaction of carriers with the laser and the recombination radiation fields respectively, and $\varepsilon^{x}_{\vec{k}} = \hbar^2k^2/2m_x$ with m_x being the exciton mass.

In Eqs. (3) the first term is the one coupling with the probe charge density, the second term is related to the change in kinetic energy in the process of polarization, the third term is due to Coulomb interaction among the carriers treated in the random phase approximation (RPA), and the last two terms are associated to relaxation effects involving mainly recombination processes. The contribution of the first term in Eq. (4) becomes negligible in the final calculation and the second leads to a coupling of the density fluctuations of electrons and holes, besides the coupling generated by the Coulomb potential. We are looking for the linear response of the system. The nonlinear contribution to Eqs. (3) have been neglected, and so too are relaxation terms due to carrier-phonon interaction that are negligible compared to those resulting from recombination effects.[7]

Solving the coupled system of Eqs. (3) to obtain $n(\vec{Q},t)$ and using the relation $\varepsilon^{-1}(\vec{Q},\omega)-1 = n(\vec{Q},t)/r_o$, we obtain for the dielectric function

$$\varepsilon(\vec{Q},\omega)/\varepsilon_o \simeq 1 - V(Q)\sum_{\vec{k}} M(\vec{k},\vec{Q};\omega)D^{-1}(\vec{k},\vec{Q},\omega) \quad , \qquad (5)$$

where

$$M(\vec{k},\vec{Q};\omega) = [(f^{e}_{\vec{k}+\vec{Q}} - f^{e}_{\vec{k}}) - (f^{h}_{\vec{k}+\vec{Q}} - f^{h}_{\vec{k}})][\hbar\omega - i(B^{e}_{\vec{k}\vec{Q}} + B^{h}_{\vec{k}\vec{Q}})] - [(f^{e}_{\vec{k}+\vec{Q}} - f^{e}_{\vec{k}})(\varepsilon^{h}_{\vec{k}+\vec{Q}} - \varepsilon^{h}_{\vec{k}}) + (f^{h}_{\vec{k}+\vec{Q}} - f^{h}_{\vec{k}})(\varepsilon^{e}_{\vec{k}+\vec{Q}} - \varepsilon^{e}_{\vec{k}})] \qquad (6a)$$

$$D(\vec{k},\vec{Q};\omega)=[i(\varepsilon^{e}_{\vec{k}+\vec{Q}}-\varepsilon^{e}_{\vec{k}}+\hbar\omega)+B^{h}_{\vec{k}\vec{Q}}]$$
$$[i(\varepsilon^{h}_{\vec{k}+\vec{Q}}-\varepsilon^{h}_{\vec{k}}-\hbar\omega-B^{e}_{\vec{k}\vec{Q}})-B^{e}_{\vec{k}\vec{Q}}]+$$
$$+B^{e}_{\vec{k}\vec{Q}}\,B^{h}_{\vec{k}\vec{Q}} \qquad (6b)$$

We use the dielectric function of Eq. (5) to calculate the electronic Raman scattering cross section

$$d^2\sigma/d\omega d\Omega \sim [1-e^{-\beta\hbar\omega}]^{-1}\ \mathrm{Im}\ \varepsilon^{-1}(\vec{Q},\omega) \qquad (7)$$

where $\hbar\omega$ is the energy transfer and $\hbar\vec{Q}$ the momentum transfer in the scattering event. The bands in the Raman spectra characterize the electronic elementary excitations.[9] Numerical calculations are performed using parameters characteristic of GaAs, low temperatures ($\beta^{-1} \ll kT_{Fe(h)}$ to allow the approximate use of step functions for Fermi-Dirac distributions) and several values of n. Figure 1 shows the case $n = 10^{16}$ cm^{-3} (obtained with $\omega_L = 2.4$ eV and $I_L = 4.5$ w/cm^2) for $Q = 10^4$; 100; and 50 cm^{-1}. Other Raman spectra for larger values of n shows a similar shape, with the two lower peaks centered at nearly 0.12 and 0.25 Qv_{Fe}. For GaAs with $Q = 10^4$ and $n = 10^{16}$ cm^{-3}, we find $Qv_{Fe} \simeq 2.9$ cm^{-1}, and for $n = 10^{18}$ cm^{-3}, $Qv_{Fe} \simeq 13.5$ cm^{-1}. Figure 2 displays the real part of the dielectric function for $Q = 10^4$ cm^{-1} and $Q = 10^2$ cm^{-1}. In these and all the other cases we analyzed the spectra is composed of four bands, of which the two lowest in frequency are peaked at frequencies that vary linearly with Q as shown by Figure 3. The position of the peaks is almost in coincidence with the frequencies that make null the real part of the dielectric function, and are identified as corresponding to scattering by what may be termed acoustic plasmons. The two other bands are one at $\Omega_{p\ell}$ (scattering by optical plasmons) in a high frequency region not shown in Fig. 3, and another in the region $0.6 \lesssim \omega/Qv_{Fe} \leq 1$. Inspection of figures 1 and 2 tells us that with decreasing values of Q the latter becomes predominant and the two acoustic branches disappear, leaving a Raman spectra with the typical form of that of a single component plasma. This suggests to consider this fourth band as due to scattering by single quasi-particles.

The straight lines in figure 3 fit the values obtained from the calculated Raman spectra (dots), and writting $\omega_{A1} = S_1Q$ and $\omega_{A2} = S_2Q$, the numerical values for the acoustic-plasmon group velocities are very near the values

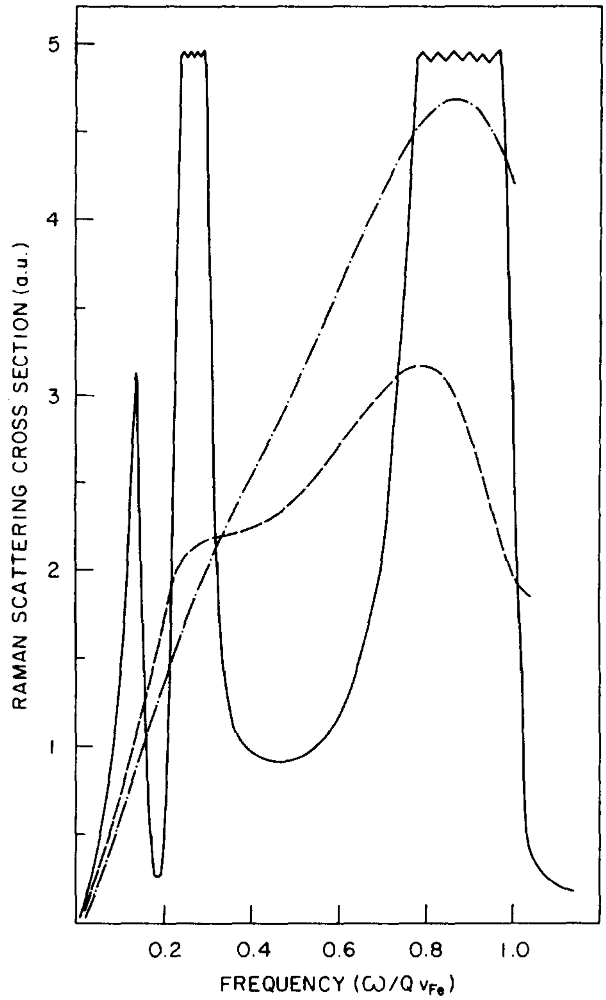

Figure 1: Raman scattering cross section. Full line (x 10^{-5}): $Q = 1\times10^4$ cm^{-1}; Dashed line (x8×10^{-6}); $Q = 100$ cm^{-1}; Dash-dotted line (x1.6×10^{-9}): $Q = 50$ cm^{-1}. The photoinjected concentration is 1×10^{16} cm^{-3}. The smearing out of all lines is visible as well as the disappearance of the low frequency ones with decreasing wavenumber.

given by

$$S_1=(m_x/3m_h)^{1/2}v_{Fe}\ ;\ S_2=(m_e m_x/3m_h^2)^{1/2}v_{Fe}, \qquad (8)$$

where v_{Fe} is the Fermi velocity of the electron. These expressions can be rewritten in the form $S_1 = \Lambda_{FT}\omega_{p\ell e}$ and $S_2 = \Lambda_{FT}\omega_{p\ell h}$, where Λ_{FT} is Fermi-Thomas screening length and $\omega_{p\ell e(h)}$ the plasma frequencies of the individual systems of electrons and of holes. This result is also analitically obtained from the expression for the unperturbed plasmon

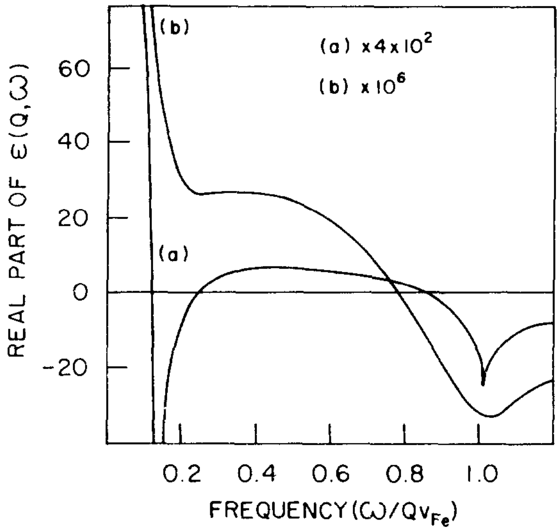

Fig. 2: Real part of the dielectric function. (a) $Q = 10^4$ cm^{-1}; (b) $Q = 100$ cm^{-1}; $n = 1 \times 10^{16}$ cm^{-3}. Two of the three zeros disappear when going from (a) to (b). Curve (b) strongly resembles that of a single component plasma.

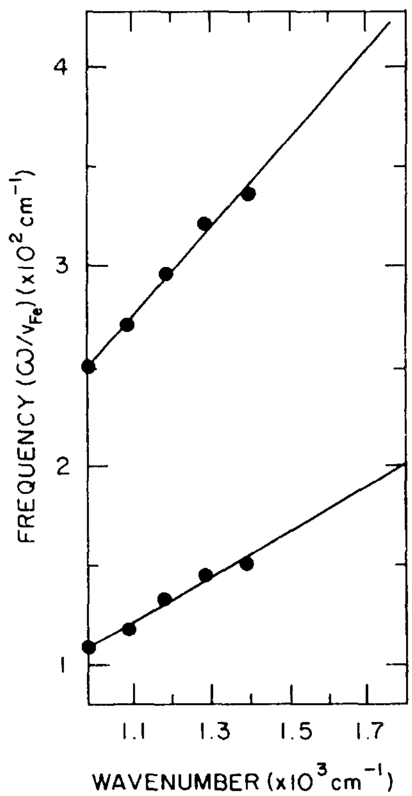

Figure 3: The acoustic plasmons dispersion relations. Dots are the positions of the peak values in the Raman spectrum; they roughly agree with the position of the first two zeros of the real part of the dielectric function.

frequency[1] $\omega^2 = (n/m)V(Q)Q^2$ if in this expression m is taken alternately as the electron and the hole mass and V is taken as the Coulomb screened potential $V^{SC}(Q) = 4\pi e^2/\varepsilon_o(Q^2+\Lambda_{FT}^{-2})$, which is approximately $4\pi e^2 \Lambda_{FT}^2/\varepsilon_o$ in the limit of small Q (The frequency of the optical plasmon, $\Omega_{p\ell}$, follows from $m \equiv m_x$ and V(Q) taken as the bare Coulomb interaction). This leads to the interpretation of the acoustic plasma waves as resulting from the collective motion of each kind of carriers interacting through the screened part of Coulomb interaction.

As already mentioned Pines and co-workers[1] suggested some time ago the existence of an acoustic plasma mode. Later on in the case of carriers in different valleys[10] and, more recently, in a study of an ideal degenerate two-component electron-hole liquid[11] this question was reinforced. Two differences can be pointed out for the case of the photoinjected carriers in nonequilibrium semiconductors with regard to the results of these authors. First, we find two instead of one acoustic branch, with group velocities satisfying the inequality $S_2 < v_{Fh} < S_1 < v_{Fe}$. This is a situation analogous to that suggested by Overhauser and Appel[12] in connection with a superconducting two-component Fermi liquid. Second, in the works of references 1, 10, and 11 the unique plasma mode is characterized as the oscillation of the heavy particles interacting through a potential screened by the light particles, while in our case each kind of carriers oscillate while interacting through a potential screened by the whole carrier system. Further, laser illumination and recombination effects play a crucial role in our analysis, in the first place to create the stationary populations of electrons and holes and next to influence and couple the charge densities of both types of carriers, reflected in coefficients B in Eqs. (3).

Our calculation is based on the random phase approximation in conjunction with the NSOM in order to describe the far-from-equilibrium state of the system. Thus, our calculation does not contain possible effects of self-energy corrections and broadening due to carrier-carrier scattering that may lead to a smearing out of the lines. It should also be remarked that the first acoustic branch, that at ω_{A2}, has a weak Raman-line intensity compared with the other lines. Further, as already noticed, both low frequency Raman lines disappear in the quasi-particle continuum Raman line for small values of Q. (The low-energy side of the latter covers the former). The results shown in the figures correspond to the case of a highly degenerate state of carriers. For carriers quasi-temperatures of a sizeable fraction of the Fermi temperature all lines are broadened and the acoustic plasmon lines disappear in the low-energy tail of the quasi-particle line. Thus, they are well defined excitations only in the highly degenerate regime of carriers, and for not too small values of the wavenumber.

In summary, we have shown that in the nonequilibrium steady state of a double component photoinjected plasma in semiconductors the electronic elementary excitation spectra is composed of the contributions from single quasi-particle excitations and three branches of collective motion, namely, the optical plasma wave associated to the relative motion of the electron-hole pairs, and two acoustic plasma waves associated with the collective motion of each of the electron and hole subsystems. These excitations should produce effects on the optical and transport (electrical and thermal) properties of the system, and therefore to its functioning in electronic devices.

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