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Free-carrier absorption by photon–ionized-impurity–plasmon processes in polar semiconductors*

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Absorption of electromagnetic radiation by free carriers is conditioned by their interaction with lattice imperfections. Both individual carrier transitions and generation of collective plasma oscillations (plasmons) may contribute to this absorption. Photon–ionized-impurity (defect)–plasmon processes at radiation frequency close to plasma frequency are discussed in detail and shown to be important in polar semiconductors with high static (lattice) dielectric constant and high carrier concentration. The theoretical results are compared with the experimental data on plasmon generation in n-PbSe in infrared.

I. INTRODUCTION

In this paper (some of the results have already been published in Ref. 1) we will consider a one-component plasma in a cubic semiconductor, consisting of free carriers (electrons or holes) of scalar, energy-independent effective mass. We will allow the existence of several equivalent band extrema for the carriers.

Suppose the magnetic field is absent, the crystal lattice is perfect and undeformed (as a consequence, all charge-charge interactions involve only the high-frequency lattice dielectric constant), and charges of ionized donors or/and acceptors are smeared out to give a uniform charge density. In such conditions, a very small dissipative part of the free-carrier dielectric function (i.e., imaginary part, corresponding to conductivity) exists only because of finite radiation wavelength.[2,3] If the radiation electric field is assumed to be uniform, as will be done in the following, the high-frequency conductivity vanishes and so does the free-carrier absorption.[4–8] The plasma response to the oscillating electric field is then purely reactive. This follows from the fact that the uniform electric field influences only the center-of-mass position and velocity but not the relative positions or velocities of the interacting carriers. Therefore it cannot excite the system of carriers. In particular, there is no contribution to free-carrier absorption from carrier-carrier or carrier-plasmon interactions, in contrary to some published calculations[9,10] in which the influence of the radiation electric field on the carrier-plasmon interaction was erroneously omitted.[6–8] The situation is not changed even by the presence of a smooth surface of the crystal, parallel to the radiation electric field.

Carrier-carrier interaction may give some free-carrier absorption in the case of energy-dependent effective mass (nonparabolic band).[5,11,12] It is well known, however, that the main contribution to free-carrier absorption is given by interactions with crystal imperfections or deformations. These interactions supply a sink for the carrier momentum and make the conservation of energy and momentum possible. Free-carrier absorption consists then in individual-carrier excitations (individual-carrier scattering), as well as in collective-carrier excitations (plasmon generation). The former contribution was studied extensively, using both classical (Drude theory) or quantum approach (see e.g., Ref. 10). In this paper we will be concerned with the latter contribution which was discussed less frequently up to now.

It should be noted that we are not interested here in magnetoplasma. We will also omit all photon–long-wavelength-plasmon processes connected with the finite dimensions of the semiconductor sample, as volume-plasmon generation at oblique incidence of light on a thin layer[13–15] or surface-plasmon generation on a rough surface.[16,17] Only the contribution of short-wavelength-plasmon generation to the high-frequency bulk conductivity (i.e. to energy dissipation) will be treated. Of course, this dissipation may, in turn, broaden the line of resonance consisting, e.g., in volume-plasmon generation in a thin layer, mentioned before.

Photon-plasmon processes were studied, as a rule, in the more general framework of photon-plasma-imperfection (deformation) interactions, in which also individual-carrier excitations were taken into account. Both nondegenerate and degenerate plasmas were considered. Usually, a theory is formulated starting from the knowledge of dielectric function for free carriers in perfect (and undeformed) crystal. For the case of plasma-ion interactions in homopolar semiconductors, a theory was developed by Ron and Tzoar[18, 19] using the analogy with gaseous plasma.[20] The contribution of photon-plasmon-ion processes to free-carrier absorption was shown to be only a small fraction of the contribution given by photon–individual-carrier–ion processes.[19] The former contribution seems to be more important in semimetals.[21] The case of plasma–optical-phonon interaction in polar semiconductors was also investigated.[12, 22] The most general formulation of the theory is due to McCumber who took into account simultaneously plasma-ion, plasma-acoustic-phonon, and plasma–optical-phonon interactions in polar semiconductors.[23] Dissipation processes involving plasmons were not, however, observed experimentally till recently.

Here we are interested only in photon–plasmon–ionized-impurity (defect) processes in polar semiconductors, and not in the contribution given by individual-carrier excitations. High-order processes involving plasmons, e.g., photon–plasmon–free-carrier–impurity processes,[7, 8, 24] are not considered. We also limit our considerations to the case of plasma frequency much higher or much lower than the LO-phonon frequency to decouple the plasmon modes from the LO modes. Because of that we can use a much simpler model of plasma and assure the physical insight in the process of plasmon generation. For the sake of definiteness we consider only electron plasma. Of course, all results hold also for hole plasma.

In Sec. II we derive a formula for the contribution to the power absorption given by photon-plasmon processes following from the interaction of plasma with an arbitrary time-independent perturbation in a nonideal (or/and deformed) crystal. In Sec. III we apply this general formula to the case of photon–plasmon–ionized-impurity (defect) processes. The result is discussed in Sec. IV and it is shown that the contribution of these processes may be comparable with the contribution from individual-carrier processes in polar semiconductors with high static lattice dielectric constant and high carrier concentration.

In the last years experimental evidence was obtained for the contribution to free-carrier absorption in the infrared given by photon–short-wavelength-plasmon processes in n-type PbSe,[25] and n-type $Pb_{1-x}Sn_xSe$.[26] In Sec. IV we compare our results with experimental findings for n-PbSe.

II. GENERAL FORMALISM FOR PLASMON GENERATION

Let us consider an electron plasma in a semiconductor, of density N_e electrons per unit volume. The number of equivalent conduction-band minima among which these electrons are distributed is w, and the minima are assumed to be spherical and parabolic.

If we want to use the weak-coupling theory of plasma, we have to assume that the mean inter-electron distance

$$r_e = (3/4\pi N_e)^{1/3} \tag{1}$$

fulfills the condition

$$r_e \lesssim a_\infty^* \tag{2}$$

for degenerate plasma, or the condition

$$r_e \lesssim r_{D\infty} \tag{3}$$

for nondegenerate (Maxwell) plasma. $a_\infty^* = \hbar^2\epsilon_\infty/e^2m^*$ is the effective Bohr radius for the electron of the charge $-e$ and effective mass m^* in the medium of the dielectric constant ϵ_∞. ϵ_∞ is the high-frequency dielectric constant of the considered crystal. $r_{D\infty} = 1/q_{D\infty}$ is the Debye screening radius for the medium of the dielectric constant ϵ_∞.

In the following the static screening of potentials due to crystal imperfections by the electron plasma will play a role. To justify the usual screening formula we have to assume

$$r_e \lesssim r_{s0} , \tag{4}$$

where r_{s0} is the Debye-Hückel screening radius with the dielectric constant of the medium taken equal to ϵ_0, the low-frequency dielectric constant of the considered crystal, i.e.,

$$q_{s0}^2 = 1/r_{s0}^2 = (4\pi e^2/\epsilon_0)(dN_e/dE_F) . \tag{5}$$

E_F is the Fermi level (measured from the bottom of the conduction band).

For nondegenerate plasma $r_{s0} = r_{D0}$, where r_{D0} is the Debye screening radius with the dielectric constant ϵ_0, i.e.,

$$q_{D0}^2 = 1/r_{D0}^2 = 4\pi e^2 N_e/\epsilon_0 k_B T . \tag{6}$$

Because of $\epsilon_0 \geq \epsilon_\infty$, condition (4) for nondegenerate plasma is weaker than condition (3). For degenerate plasma $r_{s0} = r_{\mathrm{TF}0}$, where $r_{\mathrm{TF}0}$ is the Thomas-Fermi screening radius with the dielectric constant ϵ_0, i.e.,

$$q_{\mathrm{TF}0}^2 = 1/r_{\mathrm{TF}0}^2 = (\epsilon_\infty/\epsilon_0)(12w/\pi)^{2/3}(a_\infty^* r_e)^{-1} . \tag{7}$$

Therefore, condition (4) for degenerate plasma can be written in the form

$$r_e \lesssim (\epsilon_0/\epsilon_\infty)(\pi/12w)^{2/3} a_\infty^* . \tag{8}$$

Depending on w and the ratio $\epsilon_0/\epsilon_\infty$, this condition may be weaker or stronger than condition (2).

We limit now our considerations to the case

$$\omega, \omega_{p\infty}(0) \gg \omega_{\mathrm{LO}} , \tag{9}$$

where ω and ω_{LO} are the radiation and LO-phonon frequencies, respectively. By $\omega_{p\infty}(q)$ we denote the frequency of a plasmon with wave vector $\vec{q}$ in the medium of dielectric constant ϵ_∞; therefore, $\omega_{p\infty}(0)$ is the plasma frequency

$$\omega^2_{p\infty}(0) = 4\pi e^2 N_e/\epsilon_\infty m^* . \qquad (10)$$

Under the assumption (9), plasmons are decoupled from LO phonons and plasmon frequency in the considered semiconductor is approximately $\omega_{p\infty}(q)$. If no optical phonons are present, the ionic (polar) part of the crystal polarization (i.e., this connected with the difference $\epsilon_0 - \epsilon_\infty$) and the corresponding polarization charge density $\rho(\vec{r})$ are constant in time.

As it was already mentioned, we are interested here only in collective-carrier excitations. Therefore, it is natural to use the "jelly model" to describe plasmons and their interactions with high-frequency perturbations. To justify this model, however, we have to make some assumptions. Namely, we have to assume that the excited plasma modes correspond to wave numbers q roughly equal to or smaller than both $2/r_e$ and $q_{s\infty}$, the inverse of the Debye-Hückel screening radius with the dielectric constant ϵ_∞. The former assumption is weaker than (or roughly equivalent to) the latter for nondegenerate plasma because of the already assumed condition (3). For degenerate plasma, it is also weaker (or roughly equivalent) because of condition (2), providing $w \lesssim 4$ [compare Eq. (7) with ϵ_0 replaced by ϵ_∞, i.e., the inverse of the Thomas-Fermi screening radius with the dielectric constant ϵ_∞, $q_{TF\infty}$]. Therefore, we have to assume only that

$$q \lesssim q_{s\infty} . \qquad (11)$$

In our considerations we will neglect Landau damping of the generated plasmons. In nondegenerate plasma Landau damping is weak if $q \lesssim (1/2\sqrt{3}) \times q_{D\infty}$.[27] In degenerate plasma (and for $q \lesssim 2/r_e$, which was already assumed) Landau damping is absent for $q \lesssim \omega_{p\infty}(0)/2v_F = (1/2\sqrt{3})q_{TF\infty}$, at least if $\hbar\omega_{p\infty}(0)$ is not much higher than E_F (v_F is the electron velocity at the Fermi level). Therefore for both cases we will use the condition [stronger than condition (11)]

$$q < \tfrac{1}{2} q_{s\infty} . \qquad (12)$$

We use the dispersion relation for plasmons

$$\omega^2_{p\infty}(q) = \omega^2_{p\infty}(0) + \langle v^2 \rangle q^2 , \qquad (13)$$

which follows from both Vlasov equation and the random-phase approximation (neglecting higher-order terms and the electron exchange effect[28]). $\langle v^2 \rangle$ is the mean square of electron velocity. For degenerate plasma this yields

$$\omega^2_{p\infty}(q) = \omega^2_{p\infty}(0) + \tfrac{3}{5} v_F^2 q^2 , \qquad (14)$$

and for nondegenerate plasma

$$\omega^2_{p\infty}(q) = \omega^2_{p\infty}(0) + (3k_B T/m^*) q^2 . \qquad (15)$$

Using Eqs. (14) and (15) one can show that the condition (12) is roughly equivalent to

$$[\omega_{p\infty}(q)/\omega_{p\infty}(0)] - 1 < \tfrac{1}{4} \qquad (16)$$

for both degenerate and nondegenerate plasmas. In the following, we will replace $\omega_{p\infty}(q)$ by ω in condition (16), as only plasmons with $\omega_{p\infty}(q) = \omega$ are generated by the radiation field.

The procedure of calculating high-frequency conductivity given by plasmon processes and then adding the free-carrier conductivity calculated as if the collective excitations do not exist at all, may be justified only if the ratio of the number of plasma modes to the total number of degrees of freedom of the electron system is small. This ratio is usually, in fact, of the order of 1% in metals and highly doped semiconductors.[29] Also calculating the number of plasma modes allowed by our conditions (12), (2), and (3), and dividing by $3N_e$, we obtain a ratio not exceeding a few percent or few per mil for degenerate and nondegenerate plasma, respectively.

Considering plasma dynamics it is convenient to introduce two artificial, mutually compensating uniform charge densities $+eN_e$ and $-eN_e$. Our model consists now, first of all, of an "ideal plasma" in the medium of dielectric constant ϵ_∞ [assumption (9)], i.e., of electrons and uniform charge density $+eN_e$.

There are two perturbations of this ideal plasma. The first is connected with the electron potential energy in the presence of crystal imperfections, e.g., neutral or ionized impurities, defects, dislocations, etc. (in this paper we are interested only in time-independent imperfections). The mean electric charge density of these crystal imperfections is $+eN_e$ (from the electric neutrality requirement). We denote the electron potential energy in the presence of crystal imperfections and of the uniform charge density $-eN_e$ (in the medium of dielectric constant ϵ_∞) by $U(\vec{r})$.

The second perturbation of the ideal plasma is the field given by the polarization charge density $\rho(\vec{r})$. Of course, the average of this charge density vanishes. Because of assumption (9), $\rho(\vec{r})$ is constant in time. The electron potential energy due to the charge density $\rho(\vec{r})$ (in the medium of dielectric constant ϵ_∞) will be denoted by $U_P(\vec{r})$.

As we are interested only in high-frequency conductivity due to generation of plasmons of wavelengths shorter than that of radiation, we can assume that the radiation electric field is uniform, i.e., is of the form

$$\mathrm{Re}[\vec{E}\exp(-i\omega t)]\,, \tag{17}$$

where $\vec{E}$ is a complex vector. It is then convenient to introduce a noninertial reference system ("interaction representation")[4, 19, 21, 30]

$$\vec{r}\,' = \vec{r} - (e/m^*\omega^2)\mathrm{Re}[\vec{E}\exp(-i\omega t)]\,. \tag{18}$$

In the presence of electric field (17), the motion of electrons of the ideal plasma in the noninertial reference system (18) is exactly the same as the motion in the rest reference system in the absence of radiation. In other words, in the noninertial reference system (18) the electron-radiation interaction is eliminated from the ideal-plasma Hamiltonian.

On the other hand, in the noninertial reference system the electron potential energy $U + U_P$ is no more time independent. Assuming a weak radiation field and expanding up to the linear term in $\vec{E}$, we obtain

$$\begin{aligned} U(\vec{r}\,', t) + U_P(\vec{r}\,', t) = U(\vec{r}\,') + U_P(\vec{r}\,') \\ + (e/m^*\omega^2)\mathrm{Re}[\vec{E}\exp(-i\omega t)] \\ \cdot[\nabla U(\vec{r}\,') + \nabla U_P(\vec{r}\,')]\,. \end{aligned} \tag{19}$$

The collective-excitation part of the ideal plasma Hamiltonian (in the noninertial reference system) can be written in the form

$$H_0 = {\sum_{\vec{q}}}' \hbar\omega_{p\infty}(q)(b^\dagger_{\vec{q}} b_{\vec{q}} + \tfrac{1}{2})\,, \tag{20}$$

where $b^\dagger_{\vec{q}}$ and $b_{\vec{q}}$ are the creation and annihilation operators, respectively, for the plasmon mode of the wave vector $\vec{q}$. The summation is over all plasmon modes ($\vec{q} = 0$ is excluded). The $\vec{q}$ vectors of plasmon modes are distributed in $\vec{k}$ space with the density $(2\pi)^{-3}V$, where V is the volume of the crystal (or rather of the periodicity box).

The part of plasma Hamiltonian describing the interaction of plasmon modes with crystal imperfections and polarization charges (in the noninterial reference system) is obtained as follows. Let us denote by $d(\vec{r}\,')$ the change of electron density in the point $\vec{r}\,'$ due to plasmon modes. Using the "jelly model" [and replacing $\omega_{p\infty}(0)$ by $\omega_{p\infty}(q)$] one obtains for small values of q

$$\begin{aligned} d(\vec{r}\,') = {\sum_{\vec{q}}}' [\hbar N_e/2Vm^*\omega_{p\infty}(q)]^{1/2} \\ \times q[-i\exp(i\vec{q}\cdot\vec{r}\,')b_{\vec{q}} + \mathrm{H.c.}]\,. \end{aligned} \tag{21}$$

As we are interested only in calculating radiation-induced transitions (in the lowest order), we take into account only the last term of expression (19). Based on the jelly model, we multiply it by expression (21) and integrate over V. V is assumed to be the periodicity box also for U and U_P. The resulting perturbation H' can be written in the form

$$H' = -[\hbar e^2 N_e/2Vm^{*3}\omega^4)^{1/2}\,\mathrm{Re}[\vec{E}\exp(-i\omega t)]\cdot{\sum_{\vec{q}}}' q\omega_{p\infty}^{-1/2}(q)\vec{q}\left[\left(\int_V \exp(i\vec{q}\cdot\vec{r}\,')[U(\vec{r}\,') + U_P(\vec{r}\,')]d^3r'\right)b_{\vec{q}} + \mathrm{H.c.}\right]. \tag{22}$$

We obtain the net power absorbed by the mode $\vec{q}$ being initially in the state n multiplying $\hbar\omega_{p\infty}(q)$ by the difference of transition rates for $n\to n+1$ and $n\to n-1$ transitions (only these transitions are allowed by the perturbation H'). The result is independent of n. Summing it over all modes $\vec{q}$ we obtain the total power absorption p in the volume V of the crystal, due to plasmon processes:

$$p = (\pi e^2 N_e/4Vm^{*3}\omega^4){\sum_{\vec{q}}}' \delta(\omega - \omega_{p\infty}(q))\,q^2|\vec{E}\cdot\vec{q}|^2\left|\int_V \exp(i\vec{q}\cdot\vec{r})[U(\vec{r}) + U_P(\vec{r})]\,d^3r\right|^2\,. \tag{23}$$

We have replaced $\vec{r}\,'$ by $\vec{r}$ in the integral over V. It should be noted that (for $\vec{q}\neq 0$)

$$\begin{aligned} \int_V \exp(i\vec{q}\cdot\vec{r})[U(\vec{r}) + U_P(\vec{r})]\,d^3r \\ = -q^{-2}\int_V \exp(i\vec{q}\cdot\vec{r})[\Delta U(\vec{r}) + \Delta U_P(\vec{r})]\,d^3r\,. \end{aligned} \tag{24}$$

By definition,

$$\Delta U_P(\vec{r}) = (4\pi e/\epsilon_\infty)\rho(\vec{r})\,. \tag{25}$$

III. IONIZED IMPURITIES (DEFECTS) IN POLAR SEMICONDUCTORS

In this paper we are interested only in the case of $U(\vec{r})$ given by ionized impurities or ionized point defects (and by the uniform charge density $-eN_e$). Suppose there are S types of such ions in semiconductor. $Z_l e$ and N_l ($l = 1, \ldots, S$) are the charge and concentration of the l-type ion, respectively (Z_l may be a positive or negative integer). Neutrality requirement yields the condition

$$\sum_{l=1}^{S} Z_l N_l = N_e\,. \tag{26}$$

Therefore we have

$$\Delta U(\vec{r}) = (4\pi e^2/\epsilon_\infty)\left(-N_e + \sum_{l=1}^{S} Z_l\sum_{\vec{R}_l}\delta(\vec{r} - \vec{R}_l)\right), \tag{27}$$

where the sum over $\vec{R}_l$ denotes the summation over positions of l-type ions (VN_l ions in volume V).

The polarization charge density $\rho(\vec{r})$ is

$$\rho(\vec{r})=[(\epsilon_0-\epsilon_\infty)/4\pi]\Delta\varphi_{s0}(\vec{r}) . \tag{28}$$

$\varphi_{s0}(\vec{r})$ denotes the macroscopic electric potential in the absence of radiation field and plasma oscillations, i.e., the potential produced by ionized impurities (defects) in the medium of dielectric constant ϵ_0 and in the presence of free carriers. Thus $\varphi_{s0}(\vec{r})$ is the potential $(\epsilon_\infty/\epsilon_0)U(\vec{r})/(-e)$ screened by free carriers in the medium of dielectric constant ϵ_0. This yields (for $\vec{q}\neq 0$)

$$\int_V \exp(i\vec{q}\cdot\vec{r})\varphi_{s0}(\vec{r})\,d^3r = -(\epsilon_\infty/e\epsilon_0)[1+(q_{s0}/q)^2]^{-1}\int_V \exp(i\vec{q}\cdot\vec{r})U(\vec{r})\,d^3r . \tag{29}$$

From Eqs. (28) and (29) it follows

$$\int_V \exp(i\vec{q}\cdot\vec{r})\rho(\vec{r})\,d^3r = -[\epsilon_\infty(\epsilon_0-\epsilon_\infty)/4\pi e\epsilon_0][1+(q_{s0}/q)^2]^{-1} \times\int_V \exp(i\vec{q}\cdot\vec{r})\Delta U(\vec{r})\,d^3r . \tag{30}$$

Using Eqs. (24), (25), (27), and (30), we obtain from Eq. (23)

$$p=\frac{4\pi^3e^6N_e}{V\epsilon_\infty^2 m^{*3}\omega^4}\sum_{\vec{q}}{}' \delta(\omega-\omega_{p\infty}(q))|\vec{E}\cdot(\vec{q}/q)|^2\{1-(\epsilon_0-\epsilon_\infty)\epsilon_0^{-1}[1+(q_{s0}/q)^2]^{-1}\}^2\left|\sum_{l=1}^{S} Z_l\sum_{\vec{R}_l\in V}\exp(i\vec{q}\cdot\vec{R}_l)\right|^2 . \tag{31}$$

We assume the perfectly uncorrelated and random distribution of ionized impurities (defects) in the crystal and replace the last term on the right-hand side of Eq. (31) by its average value (for $\vec{q}\neq 0$)

$$\left\langle\left|\sum_{l=1}^{S} Z_l\sum_{\vec{R}_l\in V}\exp(i\vec{q}\cdot\vec{R}_l)\right|^2\right\rangle_{\text{av}}=V\sum_{l=1}^{S} Z_l^2N_l . \tag{32}$$

Replacing now the summation over $\vec{q}$ by integration and using Eq. (13) we find

$$p=[V\epsilon_\infty\omega_{p\infty}^6(0)D/96\pi^2N_e\langle v^2\rangle^{3/2}]\,|\vec{E}|^2\omega^{-3}[\omega^2-\omega_{p\infty}^2(0)]^{1/2}(1-(\epsilon_0-\epsilon_\infty)\epsilon_0^{-1}\{1+q_{s0}^2\langle v^2\rangle[\omega^2-\omega_{p\infty}^2(0)]^{-1}\}^{-1})^2 \tag{33}$$

for $\omega>\omega_{p\infty}(0)$, and $p=0$ for $\omega\leqslant\omega_{p\infty}(0)$. D is defined

$$D=\sum_{l=1}^{S} Z_l^2(N_l/N_e) . \tag{34}$$

It depends, therefore, only on concentration ratios and ion charges. It follows from Eq. (26) that $D\geqslant 1$.

The power absorption p is related to the real part of high-frequency conductivity σ given by plasmon processes by the formula

$$p=\tfrac{1}{2}V|\vec{E}|^2\,\text{Re}\sigma . \tag{35}$$

In the Drude theory of free-carrier absorption, if

$$\omega\gg\tau^{-1} , \tag{36}$$

where τ is the momentum relaxation time, the real part of conductivity is

$$\text{Re}\sigma=[\epsilon_\infty\omega_{p\infty}^2(0)/4\pi\omega^2]\tau^{-1} . \tag{37}$$

Using Eqs. (35) and (37) we can present the formula (33) in a simpler form:

$$\tau^{-1}=[\omega_{p\infty}^4(0)D/12\pi N_e\langle v^2\rangle^{3/2}]\{1-[\omega_{p\infty}(0)/\omega]^2\}^{1/2} \times[1-[1-(\epsilon_\infty/\epsilon_0)](1+[q_{s0}^2\langle v^2\rangle/\omega_{p\infty}^2(0)]\{[\omega/\omega_{p\infty}(0)]^2-1\}^{-1})^{-1}]^2 \tag{38}$$

for $\omega>\omega_{p\infty}(0)$, and $\tau^{-1}=0$ for $\omega\leqslant\omega_{p\infty}(0)$.

It should be stressed that in our problem the relation (37) is nothing but the definition (for given Reσ or p) of a frequency-dependent, "collective" momentum-relaxation time τ corresponding to plasmon processes. Of course, it has nothing to do with some averages over energy-dependent relaxation times for individual carrier scattering. Moreover, frequency dependence of relaxation time violates the dispersion relations of Drude theory. Therefore, it would be inaccurate to use the Drude formula for Imσ with the relaxation time defined by Eq. (37).

As the real parts of high-frequency conductivities are additive, also τ^{-1} corresponding to plasmon processes is additive to those given by other momentum-relaxation processes (in particular, individual carrier scatterings).

The condition (36) is not only necessary for justifying the simple form (37) of $\mathrm{Re}\sigma$ in the Drude theory. It is also necessary for justifying our weak-perturbation treatment, as it can be seen from the following argument. The mean kinetic energy of plasma moving in the electric field (17) is

$$(Ve^2N_e/4m^*\omega^2)|\vec{E}|^2 . \tag{39}$$

The ratio of p, multiplied by the period $2\pi/\omega$, to the expression (39) should be small. From Eqs. (35) and (37), this ratio equals $4\pi/\omega\tau$, thus yielding condition (36).

It is interesting to observe that τ^{-1} given by Eq. (38) may involve the constant $\hbar$ only through $\langle v^2\rangle$ and q_{s0}^2 in the case of quantum (degenerate) statistics of electrons. In principle, therefore, our result is a classical one.

In the case of nondegenerate plasma $q_{s0}=q_{D0}$ and

$$\langle v^2\rangle = 3k_BT/m^* . \tag{40}$$

Using Eqs. (6), (10), and (40), we obtain from Eq. (38)

$$\tau^{-1}=(D/3^{7/2})\omega_{p\infty}(0)(q_{D\infty}r_e)^3\{1-[\omega_{p\infty}(0)/\omega]^2\}^{1/2}\,[1-[1-(\epsilon_\infty/\epsilon_0)](1+3(\epsilon_\infty/\epsilon_0)\{[\omega/\omega_{p\infty}(0)]^2-1\}^{-1})^{-1}]^2 , \tag{41}$$

where $q_{D\infty}$ is given by the formula (6) with ϵ_0 replaced by ϵ_∞, and r_e is given by definition (1).

In the case of degenerate plasma $q_{s0}=q_{\mathrm{TF}0}$ and

$$\langle v^2\rangle = \tfrac{3}{5}\,v_F^2 . \tag{42}$$

Using Eqs. (1), (7), (10), (42), and

$$N_e=2^{3/2}w\,m^{*3/2}E_F^{3/2}/3\pi^2\hbar^3 , \tag{43}$$

we obtain from Eq. (38)

$$\tau^{-1}=(5^{3/2}\pi D/2^5 3^{3/2}w)\omega_{p\infty}(0)[\hbar\omega_{p\infty}(0)/E_F]^3\{1-[\omega_{p\infty}(0)/\omega]^2\}^{1/2}$$
$$\times[1-[1-(\epsilon_\infty/\epsilon_0)](1+\tfrac{9}{5}(\epsilon_\infty/\epsilon_0)\{[\omega/\omega_{p\infty}(0)]^2-1\}^{-1})^{-1}]^2 . \tag{44}$$

Suppose that D is of the order of 1. It follows from assumption (3) and Eq. (41) that the condition (36) is fulfilled in the case of nondegenerate plasma. For degenerate plasma, Eq. (44) yields the same result, at least if $\hbar\omega_{p\infty}(0)$ is not much higher than E_F.

All our considerations were based on assumption (9). Let us now assume the opposite. More precisely, we assume for a while

$$\omega,\ \omega_{p0}(0)\ll\omega_{\mathrm{TO}}, \tag{45}$$

where $\omega_{p0}(0)$ is the plasma frequency in the medium of dielectric constant ϵ_0, and ω_{TO} is the TO-phonon frequency. Also under this assumption plasmonlike modes [of frequencies $\omega_{p0}(q)$] do exist. However, these plasmons are accompanied by an oscillating ionic (polar) crystal polarization. Moreover, in the presence of radiation electric field, the ionic (polar) polarization due to the screened field of ionized impurities (defects) is not constant in time. The assumptions (2) and (3) remain unchanged, and assumption (4) [i.e., assumption (8)] can be omitted. In all other considerations and results only low-frequency dielectric constant ϵ_0 appears. Therefore we have to replace throughout our calculations ϵ_∞ by ϵ_0, $\omega_{p\infty}(0)$ by $\omega_{p0}(0)$, etc., and to put $\rho(\vec{r})$ and $U_P(\vec{r})$ equal to zero.

IV. DISCUSSION AND COMPARISON WITH EXPERIMENT

Let us discuss first the dependence of D on the kind of ionized impurities (point defects) present in the crystal. As it was mentioned already, $D\geq 1$. It follows also from Eq. (26) that $D=1$ only if all $Z_l=+1$. If all $Z_l=Z$ ($Z>0$), then $D=Z$. Suppose, finally, that there are both positive and negative ionized impurities (defects) of the charges Ze ($Z>0$) and $-Ze$ and concentrations N_+ and N_-, respectively ($ZN_+-ZN_-=N_e$). Denoting

$$K=N_-/N_+ \tag{46}$$

(K is the compensation ratio if all donors and acceptors are ionized) we have

$$D=Z(1+K)/(1-K) . \tag{47}$$

For both nondegenerate [Eq. (41)] and degenerate [Eq. (44)] plasmas, changing the electron concentration N_e we change the ω scale for τ^{-1}, as it depends on the ratio $\omega/\omega_{p\infty}(0)$. The magnitude of τ^{-1} (for fixed D) is proportional to N_e in the nondegenerate case but independent of N_e in the case of degeneracy.

For given m^*, ϵ_∞, ϵ_0, D, and N_e, τ^{-1} is temperature independent for degenerate plasma, and proportional to $T^{-3/2}$ for nondegenerate plasma.

The most interesting role is played in the general expression for τ^{-1} [Eq. (38)] by q_{s0}, the inverse of the Debye-Hückel static screening radius. It is so, however, only for $\epsilon_0 > \epsilon_\infty$. If $\epsilon_0 = \epsilon_\infty$, the last factor on the right-hand side of Eq. (38) is equal identically to 1. If inequality (45) is assumed instead of (9), τ^{-1} does not involve q_{s0} for any actual value of $\epsilon_\infty/\epsilon_0$. It is clear, therefore, that the last factor of τ^{-1} is due to the time-independent ionic (polar) polarization charge density $\rho(\vec{r})$ which depends on q_{s0} [Eq. (30)]. If the last factor of τ^{-1} is dropped, $\omega \simeq \omega_{p\infty}(0)$, $w = \epsilon_\infty = 1$, $D = Z$, and m^* is the free-electron mass, our formulas (41) and (44) coincide with the results of Dawson and Oberman[20] and of Ron and Tzoar,[19,31] respectively.

It follows from Eqs. (27) and (30) that $\rho(\vec{r})$ is of the form

$$\rho(\vec{r}) = \sum_{l=1}^{S} \sum_{\vec{R}_l} Z_l \eta(\vec{r} - \vec{R}_l) , \tag{48}$$

where

$$\eta(\vec{r}) = [(\epsilon_0 - \epsilon_\infty)/\epsilon_0] \times e[-\delta(\vec{r}) + (q_{s0}^3/4\pi)(q_{s0} r)^{-1} \exp(-q_{s0} r)] . \tag{49}$$

The total charge corresponding to the charge density $\eta(\vec{r})$ vanishes.

Formula (48) means that each ionized impurity (ionized point defect) is accompanied by the charge density $Z_l \eta$ constant in time. This charge density reduces the point charge of the ion by the factor $\epsilon_\infty/\epsilon_0$, and the "substracted" charge $[(\epsilon_0 - \epsilon_\infty)/\epsilon_0] \times Z_l e$ is smeared over a volume of the radius of the order of r_{s0}. It follows that for the interacting plasmon of wavelength much larger than r_{s0} (i.e., for $q \ll q_{s0}$) the net charge density given by the ion and by $Z_l \eta$ can be approximated by a point charge $Z_l e$ as if ϵ_0 would be equal to ϵ_∞. On the other hand, if $q \gg q_{s0}$, the interacting plasmon feels only the reduced point-charge density of the ion $(\epsilon_\infty/\epsilon_0) \times Z_l e \delta(\vec{r})$. Using Eq. (13), one can observe that for $q \ll q_{s0}$ [i.e., ω close to $\omega_{p\infty}(0)$] the last factor of τ^{-1} is equal to 1, while it is $(\epsilon_\infty/\epsilon_0)^2$ for $q \gg q_{s0}$.[32] In the former case τ^{-1} involves only the high-frequency dielectric constant ϵ_∞. Paradoxically, therefore, the free-carrier screening increases τ^{-1} (for $q < q_{s0}$).

It should be noted that the inverse of relaxation time corresponding to scattering of individual electrons on ionized impurities is proportional to ϵ_0^{-2}. This type of scattering dominates usually at low temperatures. Therefore, it follows from our previous discussion that the plasmon generation may compete with individual-carrier scatterings in giving the free-carrier absorption only in polar semiconductors with $\epsilon_0 \gg \epsilon_\infty$, at rather high concentrations [cf. condition (9)], low temperatures, and high D values.

For the degenerate plasma and $\epsilon_0 \gg \epsilon_\infty$, τ^{-1} given by Eq. (44) has the maximum at

$$\omega_{\max} \cong \omega_{p\infty}(0)[1 + (3\epsilon_\infty/10\epsilon_0)] . \tag{50}$$

There is

$$\tau^{-1}(\omega_{\max}) \cong (15\pi D/2^9 w)\omega_{p\infty}(0) \times [\hbar\omega_{p\infty}(0)/E_F]^3 (\epsilon_\infty/\epsilon_0)^{1/2} , \tag{51}$$

i.e., plasmon generation decreases with increasing ϵ_0 only like $\epsilon_0^{-1/2}$.

In the last years experimental evidence was obtained for the plasmon-generation contribution to free-carrier absorption in semiconductors. It was observed, first of all, that the value of τ^{-1} measured by optical methods in n-type PbSe at low temperatures is nearly two orders of magnitude higher than the value determined from dc measurements.[33] A similar discrepancy was observed in p-PbSe,[34] and it seems to be present also in n-PbTe.[35] However, it was not observed in an n-PbSe sample with very low dc mobility, i.e., very short dc relaxation time.[36]

For n-PbSe, this effect was interpreted by inelastic scattering of individual carriers at high frequencies, in particular by optical phonons.[37-39] Then, $\tau^{-1}(\omega)$ defined as in the present paper was obtained by fitting the experimental magnetoreflectivity curves and discussed in detail.[25] $\tau^{-1}(\omega)$ seems to be a superposition of a rather flat curve which can be interpreted by individual electron–optical-phonon(s) processes, and of a bump with an edge corresponding to $\omega_{p\infty}(0)$ (Fig. 1), which may be given by plasmon-generation processes.[40] A similar shape of $\tau^{-1}(\omega)$ was observed also in n-type $Pb_{1-x}Sn_xSe$ mixed crystals.[26]

It is well known that lead chalcogenides are semiconductors with very high ϵ_0, of the order of

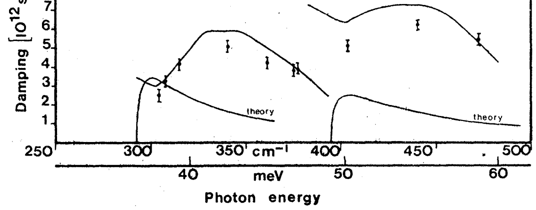

FIG. 1. Damping (τ^{-1}) in two strongly degenerated samples of n-PbSe at 30 K, as determined by two fitting methods (see Ref. 25), compared with damping calculated from Eq. (44). Samples characteristics are $N_e = 1.33$, 3.34×10^{18} cm^{-3}, $E_F = 33$ and 55 meV, and $\hbar\omega_{p\infty}(0) = 36.3$ and 48.9 meV, for samples B and C, respectively.

10^2. Therefore, one can expect the plasmon generation to be important in these materials. They do not fit well the model used in the present paper, as their four conduction-band minima (located at L points) are nonparabolic and anisotropic. Also the experiment performed in the magnetic field does not correspond well to the situation considered in this paper. Nevertheless, it is worthwhile to check if the orders of magnitude of the expected and observed effects are the same. Using the parameters $w=4$, $\epsilon_\infty=26$, $\epsilon_0=323$, and $D=7$ [as, according to Eq. (47), for $Z=1$, $K=0.75$, or $Z=2$, $K=0.56$], we have calculated τ^{-1} from Eq. (44) for two of the samples studied in Ref. 25. These theoretical plots are presented in Fig. 1. It should be noted that the dependence of the maximal theoretical value of τ^{-1} on N_e which can be seen on Fig. 1 follows from the nonparabolicity of the band, i.e., from the fact that $\omega_{p\infty}^4(0)/E_F^3$ is not independent of N_e.

In the considered material assumption (8) is weaker than assumption (2). The latter inequality is well fulfilled for both samples. As $\hbar\omega_{LO}=19$ meV, assumption (9) is rather poorly fulfilled. We have calculated the theoretical values of τ^{-1} only for the frequency range limited by the inequality (16) [in which $\omega_{p\infty}(q)$ is replaced by ω]. It should be noted that there exists evidence for twofold charged defects, at least in PbTe, and for rather high compensations.[41-43] It seems, therefore, that the used value of D is of the proper order.

It can be observed on Fig. 1 that the calculated τ^{-1} given by the photon–plasmon–ionized-impurity (defect) processes is roughly of the same magnitude as the bump on the experimental curve, for both samples. However, more accurate measurements are needed.

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