# Nonlinear Excitation of Density Fluctuations in Electron-Phonon Systems

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A nonlinear mechanism for exciting electron-phonon density fluctuation by means of a long-wavelength radiation Geld is discussed as a realistic model for doped semidonductors. Calculation of the threshold held for this phenomenon are presented, for optical as well as acoustical phonons in the infrared- and microwavefrequency regions.

## I. INTRODUCTION

**WITH** the development of high-intensity radiation sources in the microwave, infrared, and optical spectrum the study of the nonlinear interaction of electromagnetic radiation with matter stimulates considerable theoretical and experimental interest.<sup>1</sup> One area of interest is the nonlinear coupling of radiation with an electron gas. Here study of light-by-light scattering and optical mixing has been performed, assuming, however, that the electron gas can be treated as <sup>a</sup> linear system having <sup>a</sup> linearly excited mode—the plasma.<sup>2</sup> However, it was soon realized that for realistic plasmas or semiconductors, for example, the "linear approximation" assumption can not always be justified. For systems of charged particles of two species, we have two linear modes which could be coupled and excited nonlinearly via the electromagnetic field. $3-8$ 

In this paper we consider a system of interacting electrons and phonons and study, within the framework of the random-phase approximation, the excitation of resonant density fluctuations of this system; i.e., we look for excitation of plasmons and phonons driven by long-wavelength electromagnetic radiation. To see how this excitation is possible we will simply assume that the radiation field can be represented by an oscillatory homogentous electric held. In the linear approximation the field will cause the electrons to oscillate uniformly relatively to the ions in the crystal, i.e. , we impose on the electrons a uniform oscillatory velocity field. (Here we neglect the motion of the ions in the lattice, because of their large mass relatively to that of the electrons.) In the linear approximation the phonon coordinate of frequency  $\omega$  couples to electron density fluctuation at the same frequency,  $\omega$ . However, under the influence of a strong external field we also obtain nonlinear coupling to electron density fluctuations of frequencies  $\omega+n\omega_0$ ,  $n=\pm 1, \pm 2, \cdots, \omega_0$  being the frequency of the oscillating external field. Similarly, the electron density fluctuation couples, in turn, to the phonon coordinate at frequency  $\omega$  (linear effect) as well as to phonon coordinates of frequency  $\omega + n' \omega_0$ ;  $n'=\pm 1, \pm 2, \cdots$ . If, for simplicity, we assume that our system supports only two modes, the phonon mode at  $\omega$  and a plasma mode at  $\omega-\omega_0$ , we obtain a coupling mechanism in which the plasma mode at  $\omega-\omega_0$  is driven by the phonon (at frequency  $\omega$ ) and the field at frequency  $\omega_0$ , while the phonon mode (at frequency  $\omega$ ) is in turn driven by the plasma mode (at frequency  $\omega-\omega_0$  and the field (at frequency  $\omega$ ). Under these conditions the two modes will simultaneously be excited by the energy supplied by the external electric field. This can occur when tht rate of growth determined from the nonlinear mechanism overcomes the rate of loss by collisions in the system (which we take here as a macroscopic parameter).

In Sec. II we develop the general formalism of our theory, and in Sec. III we solve for realistic semiconductor the condition for parametric excitation.

#### II. GENERAL FORMALISM

Our model for a semiconductor is appropriately described by an interacting electron phonon system, whose Hamiltonian is given by

$$
H_0 = H_e + H_{ee} + H_p + H_{ep}, \tag{1a}
$$

$$
H_e = \sum_p \epsilon_p a_p^{\dagger} a_p,\tag{1b}
$$

$$
H_{ee} = \frac{1}{2} \sum_{pp'k} \varphi_k a_{p+k}{}^{\dagger} a_{p'-k}{}^{\dagger} a_{p'} a_p, \tag{1c}
$$

$$
H_p = \frac{1}{2} \sum_{k} (P_k^* P_k + \Omega_k^2 Q_k^* Q_k), \qquad (1d)
$$

$$
H_{ep} = \sum_{k} v_k Q_k \sum_{p} a_{p+k}^{\dagger} a_p. \tag{1e}
$$

Here  $a_p$ ,  $a_p^+$  are respectively the electron destruction and creation operators obeying the "Fermion" commu-518

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tation relations,  $\epsilon_p = p^2/2m$  is the kinetic energy of the electron of momentum **p**,  $\varphi_k = 4\pi e^2/k^2$  represent the electron-electron interaction,  $Q_k$  and  $P_k$  are respectively the phonon coordinate and its conjugate momentum obeying the boson commutation relations, and  $v_k$ represents the electron-phonon interaction. The response of our electron-phonon system to a homogeneous oscillatory electric field is obtained in the usual way by rewriting Eq. (1b) as

$$
H_e = \sum_p \frac{\left(\mathbf{p} - (e/c)\mathbf{A}(t)\right)^2}{2m} a_p^{\dagger} a_p. \tag{2}
$$

Here  $\mathbf{A}(t)$  is defined from the relation

$$
\mathbf{E} = -\left(1/c\right)\left(\partial \mathbf{A}/\partial t\right),\,
$$

where  $E$  is the electric field vector. We note that  $p$  in Eq.  $(1)$  and Eq.  $(2)$  is the canonical momentum. There are two terms in Eq. (2) which are field-dependent; the term proportional to  $A^2(t)$  couples to the electrons via  $\sum_{p} a_p^{\dagger} a_p = n_0$ , the average electron density and therefore does not contribute to density fluctuation of our system, and we shall disregard it. We have therefore arrived at the field-dependent Hamiltonian which is given by

$$
H = H_0 + (e/m\omega_0) \sum_p (\mathbf{p} \cdot \mathbf{E}_0(t)) a_p^{\dagger} a_p, \tag{3}
$$

with

$$
\mathbf{E}_0(t) = \mathbf{E}_0 \cos \omega_0 t, \tag{4}
$$

and the Hamiltonian  $H_0$  as defined in Eq. (1), where p is assumed to be the canonical momentum.

We next calculate the equation of motion for the one-particle density matrix which determines the coupling of the external electric field to possible density fluctuations in our electron-phonon system. We define

$$
F(p+q, p, t) = \langle a_p^+(t) a_{p+q}(t) \rangle, \tag{5}
$$

where  $\langle \rangle$  means the usual thermodynamics average over all electron coordinates and

$$
a_p(t) = \exp(iHt) a_p \exp(-iHt),
$$

etc. Using the Heisenberg picture the change in time of the operator  $a_n(t)$  is given by

$$
i(\partial/\partial t) a_p(t) = \exp(iHt) [H, a_p] \exp(-iHt), \quad (6)
$$

etc., and we therefore arrive at the following equatio of motion for the one-particle density matrix. '

$$
i(\partial/\partial t) F(p+q, p, t) + (\epsilon_p - \epsilon_{p+q}) F(p+q, p, t)
$$
  

$$
- \sum_k (\varphi_n N_k(t) + v_k Q_k(t))
$$
  

$$
\times [F(p+q-k, p, t) - F(p+q, p+k, t)]
$$
  

$$
+ (e/m\omega_0) \mathbf{q} \cdot \mathbf{E}_0 \cos \omega_0 t F(p+q, p, t) = 0. (7)
$$

 $N_q(t) = \sum_p F(p+q, p, t)$ 

 $(8)$ 

Here

and the equation of motion for the phonon coordinates is

$$
\partial^2 Q_q(t)/\partial t^2 + \Omega_q^2 Q_q = -v_q \,^* N_q(t). \tag{9}
$$

In arriving at Eq. (7) we have used the following approximations: First, electron —electron correlations (collision) have been neglected, however the selfconsistent field of the charged electrons has been taken into account. Second we consider the electron-phonon interaction only within the framework of the randomphase approximation, i.e., we neglect electron —phonon collisions and retain their interaction via the selfconsistent field only. (In the language of Feynman diagrams, we allow phonons to create or destroy electron pairs only, which represent renormalization of the polarizability due to phonons. )

We solve for  $F(p+q, p, t)$ , using a perturbation technique, however retaining  $E_0$  to all orders. We write

$$
F(p+q, p, t) = f_0(p) \, \delta_{q,0} + f_1(p, t) \, \delta_{q,0} + f(p+q, p, t), \quad (10)
$$

where  $f_0(p)$ , our zeroth-order solution, is the Fermi distribution function. In the first approximation, which is the linear response to the external field, the density matrix as well as the electric field is spatially independent and we obtain

$$
\partial f_1(p, t)/\partial t = 0;
$$
  $f_1(p, t) = f_0(p).$  (11)

The interpretation of Eq. (11) is that the first-order density matrix in terms of the canonical momentum takes the form of the Fermi distribution, and is time independent. We would like to note here that the velocity distribution of the electrons is affected by the field since

$$
m\mathbf{v} = \mathbf{p} + (e/\omega_0) \mathbf{E}_0 \cos \omega_0 t.
$$

This result is physically clear, since in the linear approximation a homogeneous field can only produce a velocity field but not density fluctuations. We next use  $f_1$  as a source for f together with the field term [in Eq.  $(7)$  which is not assumed to be small. We obtain to this order the closed set of equations:

$$
\begin{aligned}\n\left[i(\partial/\partial t) + \epsilon_p - \epsilon_{p+q}\right] f(p+q, p, t) \\
&+ (\varphi_q n_q + v_q Q_q) \left\{ f_0(p) - f_0(p+q) \right\} \\
&+ (e/m\omega_0) \mathbf{q} \cdot \mathbf{E}_0 \cos \omega_0 t f(p+q, p, t) = 0, \quad (12) \\
n_q &= \sum f(p+q, p, t), \quad (13)\n\end{aligned}
$$

$$
\partial^2 Q_q / \partial t^2 + \Omega_q{}^2 Q_q = - v_q \, {}^* n_q(t) \,. \tag{14}
$$

Only in the second-order approximation do we find a coupling to density fluctuation, which exhibits itself in the nonvanishing of  $n_q$  and the phonon coordinates  $Q_q$ . We also point out that our linearization procedure is equivalent to the random-phase approximation which one could adopt from the start. This approximation is sufhcient to describe the excitation in our electron-phonon system since we are looking only for the set of eigenmodes of the system in the presence of a strong oscillatory homogeneous electric field.

In order to solve Eqs.  $(12)$ – $(14)$  we first use a transformation which eliminates the held from our equations. Define

$$
f(p+q, p, t) = \tilde{f}(p+q, p, t) \exp(i\lambda \sin \omega_0 t), \quad (15)
$$

with

$$
\lambda = eq \cdot \mathbf{E}_0 / m \omega_0^2,
$$

we obtain a kinetic equation for  $\tilde{f}$  to be

$$
\begin{aligned} \left[ i(\partial/\partial t) + \epsilon_p - \epsilon_{p+q} \right] \tilde{f}(p+q, p, t) \\ + \varphi_q \left[ f_0(p) - f_0(p+q) \right] \rho_q(t) \end{aligned}
$$

$$
+v_q\big[ f_0(p)-f_0(p+q)\big]Q_q\exp(-i\lambda\sin\omega_0 t)=0.
$$
 (16)

Similarly the phonon coordinates obey

$$
\frac{\partial^2 Q_q}{\partial t^2} + \Omega_q^2 Q_q = -v_q * \rho_q(t) \, \exp(i\lambda \, \sin\omega_0 t), \quad (17)
$$

where

$$
\rho_q(t) = \sum_p \tilde{f}(p+q, p, t).
$$
 (18)

The physical mechanism for density excitation via the field is borne out by looking at Eq. (16). It is the difference between the oscillatory motions of the electrons and the ions (phonons) in applied fields which couples electron density fluctuations at any frequency  $\omega$  with phonon coordinates at frequencies  $\omega + s\omega_0$ ,  $s=0$   $\pm 1$ ,  $\pm 2$ , etc. (and conversely). This mode coupling is responsible for the excitation of our system. It is also clear [see Eq.  $(17)$ ] that without the retardation of the phonons no mode coupling will be possible and density fluctuation would not be exited by the field.

The solution of Eqs.  $(16-(18))$  is given in terms of the Fourier spectra of  $\tilde{f}$  and  $\rho$  where

$$
\tilde{f}(t) = \tilde{f}(\omega) \exp(-i\omega t + \eta t), \text{ etc.}
$$
 (19)

We also use the relation

$$
\exp(-i\lambda \sin \omega_0 t) = \sum_{n=-\infty}^{+\infty} J_n(\lambda) \, \exp(-in \omega_0 t), \quad (20)
$$

where  $J_n$  are the Bessel functions of the first kind. We next define

$$
P_q(\omega) = \sum_p \frac{f_0(p+q) - f_0(p)}{\epsilon_{p+q} - \epsilon_p - \omega - i\eta},
$$
\n(21)

$$
\epsilon_q(\omega) = 1 - \varphi_q P_q(\omega), \qquad (22)
$$

$$
\epsilon_q(\omega) = 1 - \varphi_q P_q(\omega), \qquad (22)
$$
  

$$
D_q(\omega) = \left[ (\omega + i\eta)^2 - \Omega_q^2 \right]^{-1}, \qquad (23)
$$

where  $P_{q\omega}$  is the electronic polarizability,  $\epsilon_{q\omega}$  the electronic dielectric function, and  $D_{\alpha\omega}$  is the phonon propagator. Solving for  $\rho_q$  in Eq. (16), we obtain

$$
\epsilon_q(\omega + s\omega_0) \rho_q(\omega + s\omega_0) = v_q P_q(\omega + s\omega_0) \times \sum_{j} J_{l-s}(\lambda) Q_q(\omega + l\omega_0).
$$
 (24)

For the phonon coordinate  $Q_q$  we solve Eq. (17) and obtain in terms of  $\rho_q$ 

$$
Q_q(\omega + s\omega_0) = v_q^* D_q(\omega + s\omega_0) \sum_l J_{l-s}(-\lambda) \rho_q(\omega + l\omega_0).
$$
\n(25)

Using Eqs. (24) and (25) we obtain after some algebra the infinite set of coupled equations;

$$
\epsilon_q(\omega + s\omega_0)\rho_q(\omega + s\omega_0) = |v_q|^2 P_q(\omega + s\omega_0)
$$
  
 
$$
\times \sum_{n,l} J_{n-s}(\lambda) J_{l-n}(-\lambda) D_q(\omega + n\omega_0)\rho_q(\omega + l\omega_0), \quad (26)
$$

which determine the eigenvalues and possible growth rates for the electron phonon system driven by the external oscillatory electric field.

Toward the end of this section we would like to check the limit  $\mathbf{E}_0 = 0$ , i.e.,  $\lambda = 0$ . In this case it is clear that only  $s=0$  component is relevant and we obtain immediately using  $J_0(0) = 1$ ,  $J_n(0) = 0$ ;  $n \neq 0$  that

$$
\left[\epsilon_q(\omega) - \mid v_q \mid ^2D_q(\omega) P_q(\omega)\right] \rho_q(\omega) = 0. \tag{27}
$$

We therefore obtain  $\lceil$  Eq. (27) $\rceil$  the dispersion relation for longitudinal oscillation for the electron phonon system in the absence of the external electric field, as expected.

#### III. NONLINEAR OPTICAL PROPERTIES OF THE ELECTRON-PHONON SYSTEM

In order to extract more useful information we consider the two-mode approximation. Here we discuss the situation in which the external field  $\mathbf{E}_0$  excites the two linear modes, the plasmon and phonon. Let us solve Eq. (26) for the case that  $\omega$  and  $\omega - \omega_0$  are respectively in the vicinity of the phonon and plasma frequencies. Ke obtain the coupled equations

$$
\epsilon_q(\omega)\rho_q(\omega)
$$
  
\n
$$
= | v_q |^2 P_q(\omega) \sum_{(n=0,-1)} \sum_{(l=0,-1)} J_n(\lambda) J_{l-n}(-\lambda)
$$
  
\n
$$
\times D_q(\omega + n\omega_0) \rho_q(\omega + l\omega_0), \quad (28)
$$
  
\n
$$
\epsilon_q(\omega - \omega_0) \rho_q(\omega - \omega_0)
$$
  
\n
$$
= | v_q |^2 P_q(\omega - \omega_0) \sum_{(n=0,-1)} \sum_{(l=0,-1)} J_{n+1}(\lambda)
$$

$$
\times J_{L-n}(-\lambda)D_q(\omega+n\omega_0)\rho_q(\omega+l\omega_0).
$$
 (29)

The solution of the coupled equations  $(28)$  and  $(29)$ , for nonvanishing density fluctuations  $\rho(\omega)$  and  $\rho(\omega-\omega_0)$ is given after much algebra by the dispersion relation

$$
\begin{split} \left[ \epsilon_q(\omega) - \left( J_0^2(\lambda) + J_1^2(\lambda) \right) \left( \mid v_q \mid^2 / \varphi_q \right) \left( 1 - \epsilon_q(\omega) \right) D_q(\omega) \right] \\ \times \left[ \epsilon_q(\omega - \omega_0) - \left( J_0^2(\lambda) + J_1^2(\lambda) \right) \left( \mid v_q \mid^2 / \varphi_q \right) \left( 1 - \epsilon_q(\omega - \omega_0) \right) D_q(\omega - \omega_0) \right] \\ + J_1^2(\lambda) \left( \mid v_q \mid^2 / \varphi_q \right) \left( \epsilon_q(\omega - \omega_0) - \epsilon_q(\omega) \right) \left( D_q(\omega) - D_q(\omega - \omega_0) \right) = 0. \end{split} \tag{30}
$$

For zero external field each of the brackets  $[\cdots]$ vhen set to be equal to zero gives respectively the dispersion relations for a plasmon or a phonon. Our solution for the nonlinear dispersion relation, Eq. (30), is correct for all systems which could be approximately described by interacting electrons and phonons, and with relaxation times small enough to preserve the identity of the linear oscillatory solutions (phonons and plasmons). However, in order to solve quantitatively Eq.  $(30)$ , we should specify our physical system. Let us choose a degenerate semiconductor in the quantum limit  $(\epsilon_F > kT)$ , to avoid contributions of electronphonon collisions to the inverse relaxation time. The plasma frequency  $\omega_p$  and the Fermi velocity  $v_F$  are

$$
\omega_p = (4\pi e^2 n / \epsilon_\infty m^*)^{1/2}, \qquad v_F = (h/m^*) (6\pi n)^{1/3}, \quad (31)
$$

where  $m^*$  is the effective mass of the electron. The Fermi-Thomas wave number  $q_{FT} = \omega_p/v_F$  which determines the limit for long-wavelength phenomena is proportional to  $n^{1/6}$ . Therefore, by changing the electron concentration by an order of magnitude we hardly effect the value of  $q_{\text{FT}}$ .

We consider semiconductors' such as InSb, InP, GaP, etc. which are ionic to a small degree and therefore provide a coupling between the longitudinal optical phonon branch and longitudinal plasma excitations. The optical phonons have a dispersion relation  $\omega_l(k) \approx$  $\omega_l(0) = \omega_l$  and their coupling to the electron gas is given in our formalism by

$$
|v_q|^2 = \varphi_q \omega_l^2 (1 - \epsilon_\infty / \epsilon_0), \qquad (32)
$$

with

$$
\varphi_q = 4\pi e^2 / \epsilon_\infty q^2. \tag{33}
$$

Here  $\epsilon_{\infty}$  is the dielectric constant of the lattice at frequency  $\omega \rightarrow \infty$  and  $\epsilon_0$  is defined from  $\omega_t^2 \epsilon_0 = \omega_l^2 \epsilon_{\infty}$ ,  $\omega_t$ being the frequency of the transverse optical mode, at long wavelengths. In the zero-wavelength case we obtain from  $\epsilon_{q=0}(\omega) = 0$  the relation

$$
1 - \left(\omega_p^2/\omega^2\right) - \left(\omega_p^2/\omega^2\right)\left(\omega_l^2/\omega^2 - \omega_l^2\right)\left(1 - \epsilon_\infty/\epsilon_0\right) = 0,\tag{34}
$$

which determines respectively the renormalized plasma frequency  $\omega_1$  and phonon frequency  $\omega_2$  given by

$$
\omega_{1,2}^{2}(0) = \frac{1}{2} (\omega_{p}^{2} + \omega_{l}^{2}) \pm \frac{1}{2} (\omega_{p}^{2} - \omega_{l}^{2})
$$
  
 
$$
\times \{1 + [4\omega_{p}^{2} \omega_{l}^{2} / (\omega_{p}^{2} - \omega_{l}^{2})^{2}] (1 - \epsilon_{\infty}/\epsilon_{0}) \}^{1/2} \quad (35)
$$

as expected. We next solve Eq.  $(30)$  for finite and large  $q$  to emphasize our effect, however, in the regime  $q\!<\!q_{\texttt{F}^{\eta}}$ and  $\omega/qv_F>1$ , and neglect the small shift in the phonon and plasmon frequency due to the external field  $\mathbf{E}_0$ . Using Eqs.  $(21)$ ,  $(22)$ , and  $(23)$  in the high-frequency, long-wavelength limit stated above, Eq. (30) reads after some algebra

$$
(\omega^2 - \omega_1^2) (\omega^2 - \omega_2^2) \left[ (\omega - \omega_0)^2 - \omega_1^2 \right] \left[ (\omega - \omega_0)^2 - \omega_2^2 \right]
$$
  
- 
$$
J_1^2(\lambda) (1 - \epsilon_\infty/\epsilon_0) \omega_1^2 \omega_2^2 (\omega_1^2 - \omega_2^2)^2 = 0, \quad (36)
$$

where we have neglected terms of order  $v_F k/\omega_l$ ,  $v_F k/\omega_p$ and already substituted in the right-hand side of Eq. (36) the linear solution, i.e.,  $\omega = \omega_1$ , and  $|\omega - \omega_0| = \omega_2$ . Here  $\omega_1$  and  $\omega_2$  are respectively the low- and high frequencies eigenvalues of the linear dispersion equation given by

$$
(\omega^2 - {\omega_l}^2) (\omega^2 - {\omega_p}^2 - \frac{3}{5}({\omega_p}^2/\omega^2) v_F^2 q^2)
$$

$$
-{\omega_p}^2 {\omega_l}^2 (1 - {\epsilon_\infty}/\epsilon_0) = 0. \quad (37)
$$

We next solve for the growth rate by retaining in the left-hand side of Eq. (36) only those terms which diverge if we substitute  $\omega = \omega_1$ . We therefore obtain by substituting  $\omega = \omega_1 + \gamma$  and assuming  $\omega_2 > \gamma$  the result

$$
\gamma^2 \left[2\omega_1 + \gamma\right] = -J_1^2(1 - \epsilon_\infty/\epsilon_0) \left(\omega_l^2 \omega_p^2 / 2\omega_2\right), \quad (38)
$$

which reduces for the case  $2\omega_1 > \gamma$  to the growth rate

$$
|\gamma| = J_1(\lambda) (1 - \epsilon_{\infty}/\epsilon_0) [\omega_l \omega_p / 2(\omega_1 \omega_2)^{1/2}]. \quad (39)
$$

In order to estimate the effect we use for  $\omega_1$  and  $\omega_2$ the solution for infinite wavelength given by Eq. (35).Wc therefore obtain for the growth rate

$$
|\gamma| \approx \frac{1}{4} \left( 1 - \frac{\epsilon_{\infty}}{\epsilon_0} \right) \frac{eqE_0}{(\epsilon_{\infty})^{1/2} m^* (\omega_1 + \omega_2)^2} \frac{\omega_i \omega_p}{(\omega_1 \omega_2)^{1/2}}.
$$
 (40)

The excitation of the density fluctuations becomes macroscopically observable when the growth rate  $\gamma$ exceeds the effective collision frequency of the system  $(\nu_1 \nu_2)^{1/2}$ , where  $\nu_1$ ,  $\nu_2$  are, respectively, the collision frequency of the two linear modes. This occurs when the radiation 6eld strength is larger than a threshold value  $(E_0)_{\text{th}}$  given by

$$
\frac{1}{4}\left(1-\frac{\epsilon_{\infty}}{\epsilon_{0}}\right)\frac{eq(E_{0})_{th}}{(\epsilon_{\infty})^{1/2}m^{*}(\omega_{1}\omega_{2})^{2}}\frac{\omega_{i}\omega_{p}}{\omega_{1}\omega_{2}} = [(\omega_{1}\tau_{1})^{1/2}(\omega_{2}\tau_{2})^{1/2}]^{-1}.
$$
\n(41)

Therefore, radiation at frequency  $\omega_0 = \omega_1 + \omega_2$  and field  $E_0$  > (E)<sub>th</sub> will exhibit an absorption. For a typical sample of InSb we obtain  $\omega_i$   $\approx$ 10<sup>13</sup>,  $\omega_p$   $\approx$ 2.10<sup>13</sup>,  $\epsilon_{\infty}/\epsilon_0 = \frac{1}{3}$ , sample of InSb we obtain  $\omega_l \approx 10^{13}$ ,  $\omega_p \approx 2.10^{13}$ ,  $\epsilon_{\infty}/\epsilon_0 = \frac{1}{3}$ ,<br> $(\epsilon_{\infty})^{1/2} = 3$ ,  $m^* = 100^{-1}m$ ,  $q = 5 \times 10^5 < q_{\text{FT}}$  and find  $\omega_1 \approx 5 \times 10^{13}$ ,  $\omega_2 \approx 0.25 \times 10^{13}$ , which we substitute in Eq. (41) to obtain

$$
(E_0)_{th}
$$
 V/cm $\approx$ 3.10<sup>5</sup> $(\omega_1 \tau_1)^{-1/2} (\omega_2 \tau_2)^{-1/2}$ . (42)

For the plasma oscillation  $\omega_1 \tau_1 \approx 10 \div 20$ . We have less accurate information about the life time of the optical phonon, and can only estimate that  $\omega_2 \approx 10 \div 100$ . We could therefore expect an absorption line at frequency  $\omega_0 = \omega_1 + \omega_2$ , for fields of the order of 10<sup>4</sup> V/cm. It is also clear that the additional absorption of the radiation and the excitation of plasma and opticalphonon fluctuations is an on-off phenomenon; no excitations are possible below  $(E_0)_{\text{th}}$  and strong excitation exist for  $E_0 \gtrsim (E_0)$ <sub>th</sub>. Therefore an experimental determination of  $(E_0)$ <sub>th</sub> allows us to determine the lifetime of the phonon provided the plasmon lifetime is known. It also should be clear that in the framework of our theory there is no way to estimate the additional

<sup>3.</sup> Hannay, Semiconductors (Reinhold Publishing Corporation, New York, 1959); J. M. Ziman. Electrons and Phonon (Clarendon Press, Oxford, England, 196Q).

absorption, i.e., the losses due to the electron-phono density excitation. However, by performing the experiment in thin films one should be able to detect the reradiation from the density fluctuation at the plasma and phonon lines, as experimental verification for their excitation.

We next calculate the threshold field for degenerate or intrinsic semiconductors in which their conduction electrons interact with the phonons via the deformation potential. The electron-phonon interaction  $v_q$  is independent of q and the parameter  $\Delta_q^2 = |v_q|^2 / \varphi_q$  is given by

$$
\Delta_q^2 = (E_k^2 K^2 / \rho_m) (q^2 / 4\pi e^2).
$$
 (43)

Here  $E_k$  is the phenomenological deformation-potential energy, **K** is the reciprocal lattice vector and  $\rho_m$  is the mass density of the crystal. The dispersion relation for long-wavelength density fluctuation is given by

$$
\epsilon_q(\omega) = 1 - (\omega_p^2/\omega^2) - (\omega_p^2/\omega^4) 3v_{\text{th}}^2 q^2 -\Delta_q^2 (\omega_p^2/\omega^2) (\omega^2 - \omega_q^2)^{-1} = 0. \quad (44)
$$

In Eq. (44) we have taken the classical limit for the dielectric function, which holds for microwave regions even for temperatures of 10'K. The solution of Eq. (30) for our system is given by

$$
(\omega^2 - \omega_1^2) (\omega^2 - \omega_2^2) \left[ (\omega - \omega_0)^2 - \omega_1^2 \right] \left[ (\omega - \omega_0)^2 - \omega_2^2 \right]
$$

$$
- J_1^2(\lambda) \Delta_q^2 \omega_p^2 (\omega_1^2 - \omega_2^2)^2 = 0, \quad (45)
$$

where  $\omega_1$  and  $\omega_2$  are the roots of Eq. (44) and are given by

$$
\omega_{1,2}^2 = \frac{1}{2} (\omega_q + \omega_p^2 + 3v_{\rm th}^2 q^2)
$$
  
 
$$
\pm \left[ \frac{1}{2} (\omega_q^2 - \omega_p^2 - 3v_{\rm th}^2 q^2)^2 + \Delta_q^2 \omega_p^2 \right]^{1/2} . \quad (46)
$$

For the case  $\Delta_q^2<\omega_q^2$ ,  $\omega_p^2$  and  $\Delta_q^2<(\omega_p-\omega_q)^2$ , we obtain the approximate solutions

$$
\omega_1^2 \le \omega_p^2 + 3v_{\rm th}^2 q^2 + 4\Delta_q^2 \omega_p^2 / (\omega_p^2 - \omega_q^2),
$$
  

$$
\omega_2^2 \le \omega_q^2 - 4\Delta_q^2 \omega_p^2 / (\omega_p^2 - \omega_q^2).
$$
 (47)

The threshold field is calculated as in the previous case, given by Eqs.  $(38)$ – $(42)$ , and we obtain

$$
\frac{1}{4} \frac{eq(E_0)_{th}}{(\epsilon_{\infty})^{1/2} m^*(\omega_1 + \omega_2)^2} \frac{\Delta_q^2 \omega_p^2}{\omega_1 \omega_2} = \left[ (\omega_1 \tau_1)^{1/2} (\omega_2 \tau_2)^{1/2} \right]^{-1} . (48)
$$

For Ge we have  $E_k \approx 18$  eV,  $K \approx 10^{+8}$  cm<sup>-1</sup>,  $\rho_m = 5.33$  $g/cm^3$ ,  $\omega_p^2 \approx 10^{21}$ ,  $\omega_q^2 \approx 10^{20}$  with  $v_{\text{th}} \approx 3 \times 10^5$  cm/sec. We obtain using Eqs.  $(43)$ ,  $(47)$ , and  $(48)$  for  $q=$ 5.10<sup>4</sup> $\lt q_p$  a threshold field

$$
(E_0)_{\text{th}} \approx 10^5 \left[ (\omega_1 \tau_1)^{1/2} (\omega_2 \tau_2)^{1/2} \right] (V/cm). \quad (49)
$$

This relatively large threshold field at microwave

frequencies is due to the poor coupling between electron and acoustical phonons for small wave numbers. For realistic case  $(E_0)$ <sub>th</sub> is of the order of 10<sup>4</sup> V/cm which can be obtained using pulse techniques.

In conclusion, we have calculated the density excitation of electron-phonon system via radiation as a realistic model for semiconductors and found the threshold field to be of the order of  $10<sup>4</sup>$  V/cm in the infrared and microwave regions.

### APPENDIX

The feasibility of the particular experiments discussed in this work depends strongly on the lifetimes of the excited modes. We would like to point out the basis for our choices of these lifetimes.

For the optical-phonon lifetime wc have used Ref. 10. Here the width of the phonon line is determined from the frequency-dependent reflectivity. The results for InSb, GaAs, etc., are,  $\omega_l \tau_l \approx 140$  at liquid-helium temperature.

Our estimate of the plasma lifetime is obtained by taking twice the single-electron lifetime  $\tau_e$  (Ref. 11) which is in turn obtained from mobility measurements (at 77°K) and the relation  $\tau_e = \mu m^*/e$ . For InAs  $(\mu=6\times10^6 \text{ cgs}, m^*\approx 0.03m, \omega_p\approx 2\times10^{13} \text{ sec}^{-1})$  and for InSb  $(\mu = 6 \times 10^7 \text{ cgs}, m^* \approx 0.016m, \omega_p \approx 2 \times 10^{13}$ for InSb  $(\mu = 6 \times 10^7 \text{ cgs}, m^* \approx 0.016m, \omega_p \approx 2 \times 10^7 \text{ sec}^{-1})$ . Hence  $\tau_e \approx 3 \times 10^{-13} \text{ sec}$  and  $\omega_p \tau_p \approx 12$ . Measure ments of the plasma lifetime for InAs has been carried recently<sup>12</sup> (similar to our conditions) and give the results:  $\nu_p \sim 100 \text{ cm}^{-1} \Delta \nu \sim 7 \text{ cm}^{-1}$ . Thus,  $\omega_p \tau_p \approx 14$ , in agreement with our estimates. We therefore conclude that our choice of  $\omega_1 \tau_1 \approx 10 \div 20$  and  $\omega_2 \tau_2 \approx 10 \div 100$  for the coupled electron —phonon system is realistic. Similar considerations for Ge with low-impurity concentration  $(\omega_p^2 \sim 10^{21})$  also show  $\omega_1 \tau_1 \approx 10$  and  $\omega_2 \tau_2 = 10 \div 100$  to be a realistic choice. We would like finally to point out that our general eigenvalue equation (26) indicates a possible density fluctuation not only at  $\omega_2$  and  $\omega_1 =$  $\omega_0-\omega_2$  but also at  $\omega_1'=\omega_0+\omega_2$ . However, as one can easily check, the mode at  $\omega_1'$  can only exist if the mode at  $\omega_1$  is excited. Therefore, the mode at  $\omega_1'$  can only increase the threshold field. In order to eliminate the mode at  $\omega_1'$ , we need a resonant mode at  $\omega_1$  such that  $2\tau_1\omega_2\gtrsim1$ , condition that where met in our work. For the case of  $2\tau \omega_2 \leq 1$  the real threshold would be somewhat higher than the one calculated here. However our result will not be applicable for  $2\tau \omega_2 \ll 1$ , and we have to extend, in this case, our analysis to include all three modes.

<sup>&</sup>lt;sup>10</sup> M. Hass and B. W. Henvis, J. Phys. Chem. Solids 23, 1099 (1962).  $"$  <sup>11</sup> H. Frohlich and H. Pelzer, Proc. Phys. Soc. (London)  $A68$ ,

<sup>525 (1955).&</sup>lt;br><sup>12</sup> C. K. N. Patel and R. E. Slusher (to be published)