Theory of Indirect Excitation of Sound by Light*

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(Received 17 June 1971)

A semiclassical theory of the excitation by light of crystalline quasiparticles coupled "indirectly" to light via an intermediate field is presented and illustrated via a model photon-exciton-phonon system. The resonance behavior of the excitation efficiency is investigated for various values of the couplings, and dependences on boundary conditions and damping effects are illustrated. Sharply resonant behavior results for sufficiently strong couplings and small enough damping, suggesting the possibility of applications of "indirect" excitation by light in actual devices, as well as in the study of microscopic properties of materials.

I. INTRODUCTION

Many of the quasiparticle fields in crystals, such as the LO- and acoustic-phonon fields,[1] do not couple directly to the photon (radiation) field. Also, even for those fields which do, in general, couple to photons (e.g., TO phonons[1,2]), the coupling constants often vanish in particular cases—for example, for symmetry reasons.[3] Since today's lasers provide us with a tunable source of intense monochromatic light, special interest attaches to whether such "inactive" modes (i.e., not coupled directly to light) may nevertheless be excited "indirectly" by an external light source. The latter is clearly possible, in principle, whenever light couples to some intermediate field, which in turn couples to the light-inactive field. Moreover, such excitation can be resonant whenever the dispersions of both pairs of fields (light-intermediate and intermediate-inactive) cross within a single narrow frequency region (roughly of width less than the smaller of the two coupling constants for the pairs). Equivalently, the dispersion of the composite field formed by the interaction of light with the intermediate field needs to cross the inactive field dispersion to lead to resonant behavior. It is the purpose of the present paper to study resonance excitation by light in such situations.

We point out some examples of systems where "indirect" excitation by photons may be of interest. (We will here always employ the terms "direct" and "indirect" with reference to the microscopic interactions between the uncoupled fields of the system.)

(a) In the presence of a magnetic field, microwave radiation incident on a metal forms a propagating composite quasiparticle called a helicon.[4,5] Although the externally incident microwaves do not couple directly to phonons, the mediation of the electron-phonon interaction provides a mechanism by which sound may be excited in this system.[5]

(b) Again, mediation by the electron-phonon interaction may allow excitation of LO phonons in semiconductors by an external light source. A possible mechanism is the excitation by the incident light of plasmons,[6] which in turn couple to the LO phonons.

(c) Should either the magnon[7] or phonon field in an insulator be light inactive, it might nevertheless be excitable whenever the magnon-phonon interaction acts in conjunction with either a phonon-photon[2] or magnon-photon[8] interaction, respectively.

(d) In narrow-gap semiconductors phonons might be excited via various intermediates. The presence of excitons and the exciton-phonon interaction[9] should allow for excitation of acoustic or optical phonons. Interaction of light with free carriers can excite plasmons[6] which may, in turn, excite LO phonons.

All the above-described systems are similar in that three fields are involved: external radiation, an intermediate field, and an "indirectly" coupled field which one desires to excite. Rather than concern ourselves with the abundant details surrounding such systems, we wish here to investigate just a single simplified model of such a system: one consisting of photons, excitons, and acoustic phonons, where the exciton and phonon dispersions cross. Such a situation is realizable, in principle, in a narrow-gap semiconductor. The main advantage of the present system is, however, that it is on the whole free of unessential complications yet general enough to encompass the principal physical and mathematical considerations involved in treating the indirect-excitation–resonance problem.

We note that although it has been demonstrated that excitons may, in principle, exist in such materials,[10] their positive identification seems to have been thwarted by the presence of high concentrations of intrinsic and/or extrinsic carriers which tend to screen out electron-hole interactions.[11] As such, the particular process of photon-induced excitation of phonons in the model system chosen

here may not yet be an experimentally feasible one.

To describe the present resonance system theoretically, we adopt polariton (composite-quasiparticle) theory,[12] an approach which is especially useful in the immediate dispersion cross-over region, where conventional perturbation theory breaks down. We employ the term "polariton" to refer to the coupled modes of the present interacting system. In our development, we formulate classical field equations and obtain the energy density, in analogy with the Born-Huang (BH) treatment of the TO-phonon–light problem.[2] We then identify the fraction of acoustic-phonon energy associated with a given polariton mode, and define an efficiency function C for sound excitation. After specifying the boundary conditions (b. c.) on the phonons,[13] we evaluate C explicitly, illustrate its frequency dependence for various values of the system parameters, and discuss the results.

II. FIELD EQUATIONS, ENERGY DENSITY, AND POLARITON DISPERSIONS

We consider an acoustic-phonon field ($\vec{V}$), exciton field ($\vec{W}$), and transverse electric field ($\vec{\mathcal{E}}$), with natural frequencies ω_0, E, and kc, respectively. Let $\vec{P}$ denote the polarization field, $\vec{\mathcal{H}}$ the magnetic field, γ the phonon-exciton bilinear coupling function, b_{22} the background dielectric susceptibility [$\equiv(\epsilon_b-1)/4\pi$, where ϵ_b is the background dielectric constant], and b_{12} the photon-exciton bilinear coupling. We set up field equations[2] for the $\vec{k}$-space Fourier components of the fields; in analogy with BH these take the form

$$\ddot{\vec{V}}=-\omega_0^2\vec{V}+\gamma\vec{W};\quad \ddot{\vec{W}}=-E^2\vec{W}+b_{12}\vec{\mathcal{E}}+\gamma\vec{V};$$
$$\vec{P}=b_{12}\vec{W}+b_{22}\vec{\mathcal{E}};\quad \vec{k}\cdot(\vec{\mathcal{E}}+4\pi\vec{P})=0;\quad \vec{k}\cdot\vec{\mathcal{H}}=0; \tag{1}$$
$$i\vec{k}\times\vec{\mathcal{E}}=-c^{-1}\dot{\vec{\mathcal{H}}};\quad i\vec{k}\times\vec{\mathcal{H}}=c^{-1}(\dot{\vec{\mathcal{E}}}+4\pi\dot{\vec{P}}).$$

In analogy with BH, we choose for the energy density,

$$U=\tfrac{1}{2}(\dot{V}^2+\omega_0^2V^2)-\gamma\vec{V}\cdot\vec{W}+\tfrac{1}{2}(\dot{W}^2+E^2W^2)-b_{12}\vec{W}\cdot\vec{\mathcal{E}}$$
$$-\tfrac{1}{2}b_{22}\mathcal{E}^2+\vec{\mathcal{E}}\cdot\vec{P}+(\mathcal{E}^2+\mathcal{H}^2)/8\pi. \tag{2}$$

This choice implies

$$\frac{dU}{dt}=\dot{\vec{P}}\cdot\vec{\mathcal{E}}+\frac{d[(\mathcal{E}^2+\mathcal{H}^2)/8\pi]}{dt}, \tag{3}$$

which may be interpreted similarly to BH: The rate of change of energy density equals the Joule heating plus the rate of change of electromagnetic field energy density.

Employing Eqs. (1) and (2) we may express U as

$$U=\tfrac{1}{2}(\dot{V}^2+\omega_0^2V^2)-\gamma\vec{V}\cdot\vec{W}+\tfrac{1}{2}(\dot{W}^2+E^2W^2)$$
$$+(\epsilon_b\mathcal{E}^2+\mathcal{H}^2)/8\pi. \tag{4}$$

Specializing to harmonic fields with time variation $e^{-i\omega t}$, we now take U to refer to the time-averaged energy density. With this redefinition, and allowing the various field variables to refer to their time-independent amplitudes, one obtains the transverse-mode[2] density U from Eqs. (1) and (4) as

$$U=\frac{\mathcal{E}^2}{4\pi}\left(\frac{[E^2-\gamma^2(\omega_0^2-2\omega^2)/(\omega_0^2-\omega^2)^2]\,4\pi b_{12}^2}{[E^2-\omega^2-\gamma^2/(\omega_0^2-\omega^2)]^2}+\epsilon_b\right). \tag{5}$$

This result reduces to a form similar to Loudon's[14] for the case $\gamma=0$. The relations between fields employed in obtaining Eq. (5) are

$$V=\frac{\gamma W}{(\omega_0^2-\omega^2)},\quad W=\frac{b_{12}\mathcal{E}}{[E^2-\omega^2-\gamma^2/(\omega_0^2-\omega^2)]}. \tag{6}$$

The polariton eigenvalue equation following from (1) is

$$\epsilon(k,\omega)\equiv\frac{k^2c^2}{\omega^2}=\epsilon_b+\frac{4\pi b_{12}^2}{[E^2-\omega^2-\gamma^2/(\omega_0^2-\omega^2)]}, \tag{7}$$

where ϵ is the dielectric function. To calculate the polariton dispersion explicitly, one must specify the k dependence of γ, E, and ω_0. For large effective mass, E is nearly independent of k for a large range in k: This approximation is adequate for our purposes. We assume that the phonon frequency ω_0 is linear in k in the region of interest here ($\omega_0=v_s k$) and take γ constant in this region.[15] We note that the present eigenvalue equation then becomes a quadratic in k^2, and that the two solutions of absolute value k_1 and k_2 specify the two degenerate polariton modes at any frequency ω. The polariton dispersions arising from Eq. (7) are illustrated for typical parameters in Fig. 1.

III. PHONON-EXCITATION EFFICIENCY

In a resonantly coupled system there is no unique unambiguous way to divide up and classify the energy according to the noninteracting fields of the system.[16] Strictly speaking, an expression for U and relations between fields [for example, such as in Eqs. (1) and (2)] represent a complete description of the interacting system. Thus, the classification of the energy according to the noninteracting fields is carried out primarily for purposes of interpretation. For such a division to be useful the portions identified in the present problem as photon, exciton, or phonon must conform to various requirements consistent with the physical properties characterizing these fields. The exact nature of these requirements will be made evident in the development which follows.

Let us first consider a simplified problem of an interacting phonon-exciton system described by density

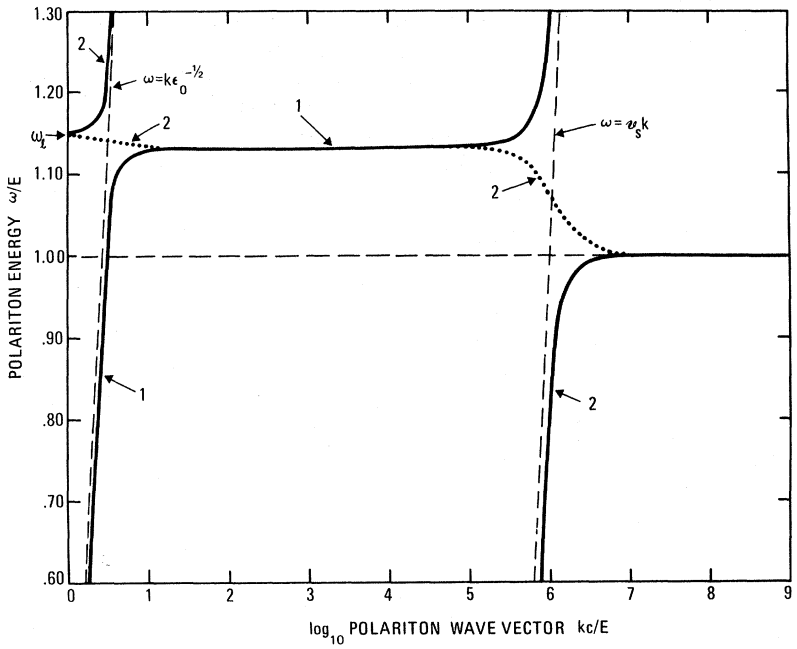

FIG. 1. Polariton dispersion relation for $A_1 = 0.4$, $B_1 = 0.36$ (cf. Ref. 22), $v_s/c \sim 10^{-6}$, $\epsilon_{b,1} = 8$, and zero damping. The polariton dispersion for real k is given by the solid lines; the dispersion for pure imaginary values of absolute value k is given by dotted lines. The dashed lines indicate the three noninteracting dispersions described in the text.

$$U = \tfrac{1}{2}(\dot{V}^2 + \omega_0^2 V^2) - \gamma \vec{\nabla} \cdot \vec{W} + \tfrac{1}{2}(\dot{W}^2 + E^2 W^2) \ . \qquad (8)$$

It will be most useful for our purposes to introduce the quantum Hamiltonian H corresponding to U, which may be shown to be[17] ($\hbar = 1$)

$$H = \sum_{\vec{k}} \{\omega_0(k) a_{\vec{k}}^{+} a_{\vec{k}}^{-} + [(\gamma/2\omega_0^{1/2} E^{1/2})(a_{\vec{k}}^{+} b_{-\vec{k}}^{+} - a_{\vec{k}}^{+} b_{-\vec{k}}^{-}) + \text{H.c.}] + E b_{\vec{k}}^{+} b_{\vec{k}}^{-}\} \ , \qquad (9)$$

where $a^{\pm}$ and $b^{\pm}$ are creation-annihilation operators for phonons and excitons, respectivity, and H.c. indicates Hermitian conjugate. One can show that the ith polariton operator A_i^{+} is a linear combination of a's and b's of the form[17,18]

$$A_{i\vec{k}}^{+} = c_{1i} a_{\vec{k}}^{+} + c_{2i} b_{\vec{k}}^{+} + d_{1i} a_{-\vec{k}}^{-} + d_{2i} b_{-\vec{k}}^{-} \ , \qquad (10)$$

where

$$c_{1i} = (i)^i (\omega_0^2 - \omega_i^2)(E + \omega_i)(4E\omega_i b_i)^{-1/2} \ ,$$
$$c_{2i} = -(i)^i \gamma (\omega_0 + \omega_i)(4\omega_i \omega_0 b_i)^{-1/2} , \qquad (11)$$
$$b_i = (\omega_0^2 - \omega_i^2)^2 + \gamma^2 \ ,$$

d_{1i} is obtained by replacing $E + \omega_i$ in c_{1i} by $-(E - \omega_i)$, and d_{2i} by replacing $\omega_0 + \omega_i$ in c_{2i} by $\omega_0 - \omega_i$. ω_1 and ω_2 are the two polariton frequencies at a given k, and $\omega_3 = -\omega_1$, $\omega_4 = -\omega_2$. The linear transformation described by Eqs. (10) and (11) suggests the definition of a probability of phonon creation via A_i^{+} as $g_i = |c_{1i}|^2 - |d_{1i}|^2$ (where $|c|^2$ represents phonon creation and $|d|^2$ phonon destruction), and an analogous exciton probability as $g_i' = |c_{2i}|^2 - |d_{2i}|^2$; then

$$g_i = \gamma^2/b_i \ , \quad g_i' = (\omega_0^2 - \omega_i^2)/b_i \ . \qquad (12)$$

We will employ the notation g_i interchangeably to refer to either fixed k [which then determines a set of $\omega_i(k)$'s] or at fixed ω [which then determines a set of $k_i(\omega)$'s]. In support of the interpretation[18] given here to the g's, we note that for either fixed k or ω the following properties hold: (i) g_i, $g_i' \leq 1$; $g_i + g_i' = 1$. (ii) At both $\omega \ll E$ and $\omega \gg E$, where the polariton dispersion is very nearly phononlike, $g_i \approx 1$, $g_i' \approx 0$. For $\omega \to E$, where the dispersion becomes very nearly excitonlike, $g_i \approx 0$, $g_i' \approx 1$.

For a given mode i we then identify the portion of energy associated with phonons as $U_1 = gU$ and with excitons as $U_2 = g'U$. As a function of frequency ω, the energy is thus entirely of phonon character for $\omega \ll E$, becoming entirely excitonlike for $\omega \to E$, and reverting again to pure phonon for $\omega \gg E$.

We may now inquire as to what division of U corresponds to the present choices for the g's. Consider the simplest conceivable division, namely, splitting the interaction energy $-\gamma \vec{\nabla} \cdot \vec{W}$ equally between the noninteracting fields; i.e.,

$$U_1 = \tfrac{1}{2}(\dot{V}^2 + \omega_0^2 V^2) - \tfrac{1}{2}\gamma \vec{\nabla} \cdot \vec{W} \ ,$$
$$U_2 = \tfrac{1}{2}(\dot{W}^2 + E^2 W^2) - \tfrac{1}{2}\gamma \vec{\nabla} \cdot \vec{W} \ . \qquad (13)$$

One easily shows that these choices for U_1 and U_2 satisfy

$$U_1/U = g, \quad U_2/U = g' \ . \qquad (14)$$

Thus it is an equal division of the interaction energy between the two fields which corresponds to the interpretation of the g's following from the quantum-transformation coefficients introduced in Eqs. (10)

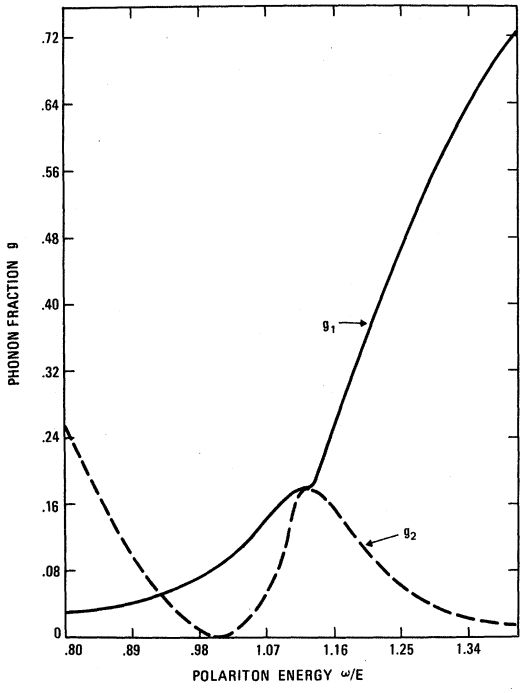

FIG. 2. Phonon fractions g_i vs ω/E for parameters of Fig. 1. The indices 1, 2, refer to the curves in Fig. 1, the solid line for 1 and the dashed line for 2.

and (11).

While the problem of an external $e''m$ field interacting with just a single other field may be formulated in a nearly analogous fashion, the three-field problem involves various complications in the quantum formulation not present in the above example (proper sum rules need be obeyed to correctly determine the coefficient of the A^2 term in the vector potential). We therefore do not attempt a quantum formulation for the present three-field problem. Rather, we note that the quantity of principal concern here is just the fraction of phonon energy in a system mode. The fact that the phonon is here coupled directly to just the exciton field alone suggests the identical division of the interaction energy $-\gamma\vec{\dot{V}}\cdot\vec{W}$ as in the absence of the $e''m$ field. Again, such an identification is useful only if the results conform to the sort of requirements satisfied above in the two-field case. With the proposed division one finds for $g_i(\omega)$

$$
\begin{aligned}
&g_i = \tfrac{1}{2}[\dot{V}^2 + \omega_0^2(i)V^2 - \gamma\vec{\dot{V}}\cdot\vec{W}] = \mu(\omega\gamma)^2(D_i^2+Q_i)^{-1},\\
&\mu = 4\pi b_{12}^2\epsilon_b^{-1},\\
&D_i = (E^2-\omega^2)[\omega_0^2(i)-\omega^2]-\gamma^2,\\
&Q_i = \mu\{E^2[\omega_0^2(i)-\omega^2]^2-\gamma^2[\omega_0^2(i)-\omega^2]+\gamma^2\omega^2\},
\end{aligned}
\tag{15}
$$

where the notation $\omega_0(i)$ indicates that we determine $k_i = k_i(\omega)$ in evaluating ω_0. One easily verifies from (15) that, as necessary, $g_i \leq 1$; for $i=1$ and $\omega \ll E$, where the polariton dispersion becomes photonlike (see Fig. 1), $g_1 \approx 0$, while for $i=2$, where the dispersion becomes phononlike, $g_2 \approx 1$. For $\omega \to E$, curve 2 becomes nearly excitonlike and $g_2 \approx 0$. Immediately above $\omega = E$, curve 2 again takes on some phonon character and g_2 increases correspondingly. For $\omega \gg E$, polariton 2 eventually becomes purely photonlike and $g_2 \approx 0$. In the same region polariton 1 becomes purely phononlike, so that $g_1 \approx 1$. These properties justify the choice of (15) for g as physically appropriate. The various properties discussed are illustrated for the crossover region, in Fig. 2 (for the same parameters as in Fig. 1), which demonstrates the frequency dependence of g_1 and g_2 in this region. Although not specifically of interest here, one can show, employing arguments similar to the above, that the exciton-energy fraction g' and the $e''m$ fraction g'' take the forms

$$
\begin{aligned}
&g' = D_i[\tfrac{1}{2}\mu(\omega_0^2-\omega^2)+D_i](D_i^2+Q_i)^{-1};\\
&g'' = \tfrac{1}{2}\mu(\omega_0^2-\omega^2)[(E^2+\omega^2)(\omega_0^2-\omega^2)-\gamma^2](D_i^2+Q_i)^{-1}.
\end{aligned}
\tag{16}
$$

These functions have properties exactly analogous to those of g.

To describe externally induced excitations in a multimode system such as the present one, we need, in addition to the fractions g, the probabilities with which each polariton is excited at the boundary.[19] We define probabilities $p_{1,2}$ that an incident photon excites polariton 1, 2; if we for the moment disregard the reflectivity, and normalize $p_1+p_2=1$, then the total phonon, or sound, energy fraction may be defined as

$$
F(\omega) = p_1(\omega)g(1) + p_2(\omega)g(2), \tag{17}
$$

where 1 and 2 in the argument of g indicate evaluation at k_1 and k_2, respectively. Incorporating the reflectivity $R(\omega)$, we may define the efficiency for sound excitation by an externally incident photon as

$$
C(\omega) \equiv [1-R(\omega)]F(\omega). \tag{18}
$$

C is interpreted as the fraction of incident photon energy which is transformed into sound energy at the crystal boundary.

To obtain F or C explicitly we must specify the relation between p_1 and p_2, which follows once the b.c.[14] on the phonons are specified for a given geometry. For normal incidence in a semiinfinite medium, a simple but reasonable b.c. is vanishing stress at the boundary.[20] If z is the coordinate normal to the surface, then we require

$$
\left(\frac{\partial V_1}{\partial z}\right)_0 + \left(\frac{\partial V_2}{\partial z}\right)_0 = 0. \tag{19}
$$

Note that this b.c. specifies $R(\omega)$ uniquely as well.

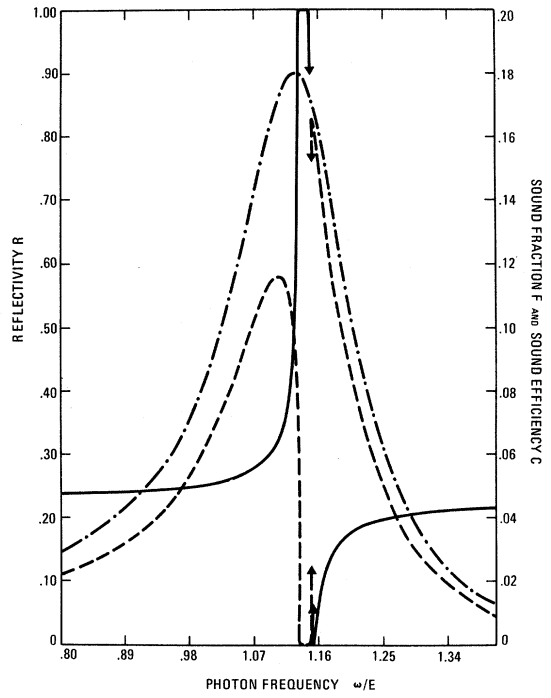

FIG. 3. Sound fraction F, sound efficiency C, and reflectivity R vs ω/E for parameters of Fig. 1. Solid line: R; dashed line: C; dash-dot line: F.

Incorporating (19), one obtains for F and R (suppressing ω for notational simplicity)

$$F = g(1) + [g(2) - g(1)](1 + p_1/p_2)^{-1}\,,$$
$$R = \left| \frac{(n_1 - 1) + (n_2 - 1)(\mathcal{E}_2/\mathcal{E}_1)}{(n_1 + 1) + (n_2 + 1)(\mathcal{E}_2/\mathcal{E}_1)} \right|^2 , \tag{20}$$

where

$$\frac{p_1}{p_2} = \frac{g(2)}{g(1)} \frac{D_2^2}{D_1^2} \left| \frac{\mathcal{E}_1}{\mathcal{E}_2} \right|^2 ; \quad \frac{\mathcal{E}_2}{\mathcal{E}_1} = -\frac{D_2 n_1}{D_1 n_2}\,,$$
$$n_i = k_i c \omega^{-1} . \tag{21}$$

For other geometries, or under otherwise more general conditions, one may derive suitably modified expressions.

Because polariton 2 is totally damped for $E < \omega < \omega_t$, we adopt the convention $p_2 = 0$ for this region. In fact, for the present case of zero damping the exact value of p_2 has a negligible effect on the value of C in this region since $1 - R$ is very nearly zero throughout.

IV. BEHAVIOR OF SOUND EXCITATION EFFICIENCY

We proceed to illustrate the frequency dependence of C for various values of the parameters of the theory, namely, γ, $\beta\,(\equiv 2\pi^{1/2} E^{1/2} b_{12})$, and ϵ_b. For an $n = 1$ hydrogenic exciton level, for example, γ and β are of the form[21] ($\hbar = 1$)

$$\gamma(\omega_0 E)^{-1/2} \equiv \gamma' = D V_0^{1/2} (2M\omega_0 a_0^3 a^2)^{-1/2}\,,$$
$$\beta = (e/m) 2^{1/2} P (\epsilon_b a_0^3)^{-1/2}\,, \tag{22}$$

where D is a deformation potential and P a momentum matrix element between Bloch states of the crystal; a_0 is the exciton Bohr radius and a the lattice constant; various other standard symbols appearing in (22) are defined in Ref. 21.

The functions F, C, and R are illustrated in Fig. 3 for values of the parameters indicated.[22] In the zero-damping model as presented above, the reflectivity is nearly perfect in the region immediately above $\omega = E$ so that C nearly vanishes in this region. Because of the dissimilar variations of F and $1 - R$, the maximum in C is more strongly peaked, but smaller in height, than the maximum in F. Also, note that one obtains for C a doubly peaked structure as a function of ω, with a lower broader peak on the low-energy side of $\omega = E$.

Figures 4–6 indicate the frequency dependence of C upon variation of γ, β, and ϵ_b, respectively. The peak positions are seen to be shifted substantially by variation of γ; also, the height and width of the peaks change substantially with variation of any of the three parameters, as is to be expected.

We will now consider two other factors relevant to the computations, namely, the effect of boundary conditions, and the effects of damping (dissipation).[2]

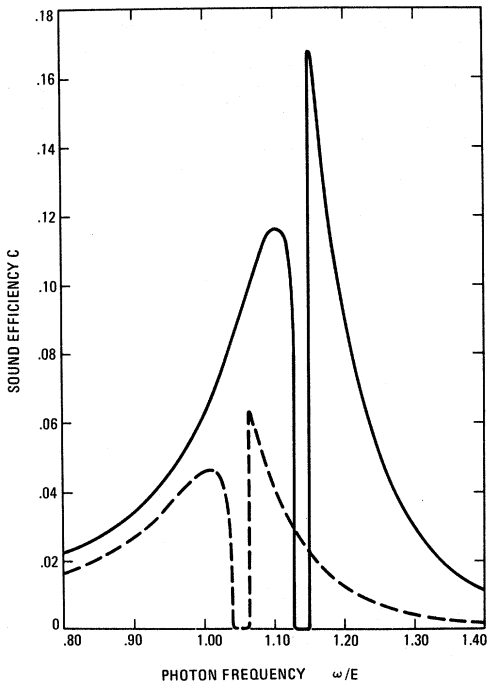

FIG. 4. Dependence of sound efficiency C vs ω/E on phonon-exciton coupling B (see Ref. 22). Solid line: $B = B_1$; dashed line: $B = 0.25 B_1$; all other parameters as in Fig. 1. Sub-1 indicates Fig. 1 values.

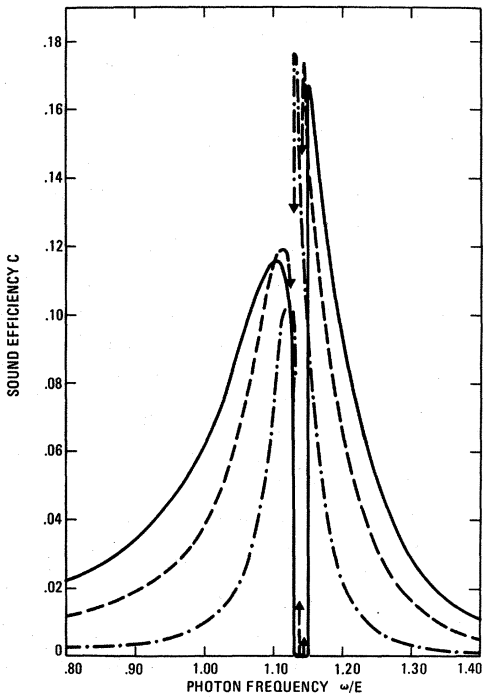

FIG. 5. Dependence of sound efficiency C vs ω/E on photon-exciton coupling A (see Ref. 22). Solid line: $A = A_1$; dashed line: $A = 0.5A_1$; dash-dot line: $A = 0.1A_1$; all other parameters as in Fig. 1. To avoid clutter in the figure we omit nearly vertical portions of the curves, indicating them instead by arrows.

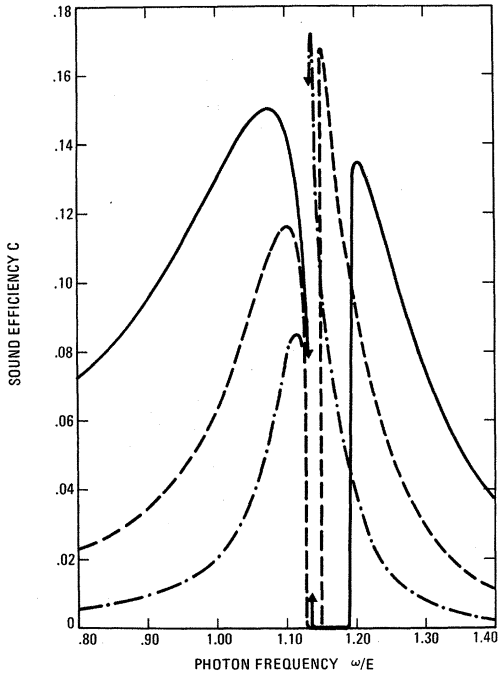

FIG. 6. Dependence of sound efficiency C vs ω/E on background dielectric constant ϵ_b. Solid line: $\epsilon_b = 0.3\ \epsilon_{b,1}$; dashed line: $\epsilon_b = \epsilon_{b,1}$; dash-dot line: $\epsilon_b = 3\epsilon_{b,1}$; all other parameters as in Fig. 1.

The b.c. employed in obtaining Figs 2–6, as given by Eq. (19), is appropriate to a free surface and for waves normal to the surface. To investigate the effect of a change in b.c., we consider the hypothetical case of a totally "clamped" surface. In this case the total displacement is made to vanish at the surface[23] and one obtains the results indicated in Fig. 7. One observes from the figure that the "clamped" case leads to a higher maximum in C; in addition, a subsidiary peak in F and C appears on the low-energy side of $\omega = E$.

The theory we have presented can be modified

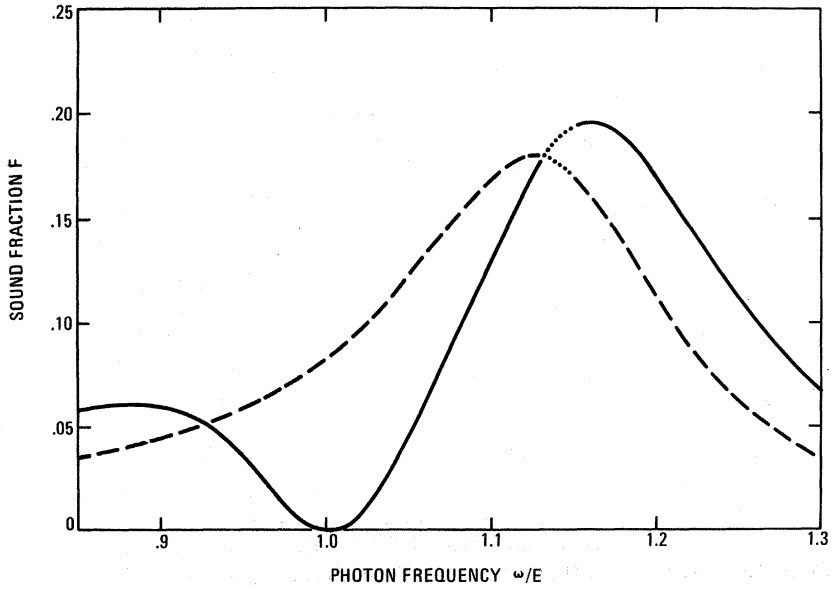

Fig. 7. Sound fraction F vs ω/E for various b.c. Solid line: $(V_1 + V_2)_0 = 0$; dashed line: $(\partial V_1/\partial z + \partial V_2/\partial z)_0 = 0$; all parameters are as in Fig. 1. The dotted portion of the curves indicates the region where k is pure imaginary.

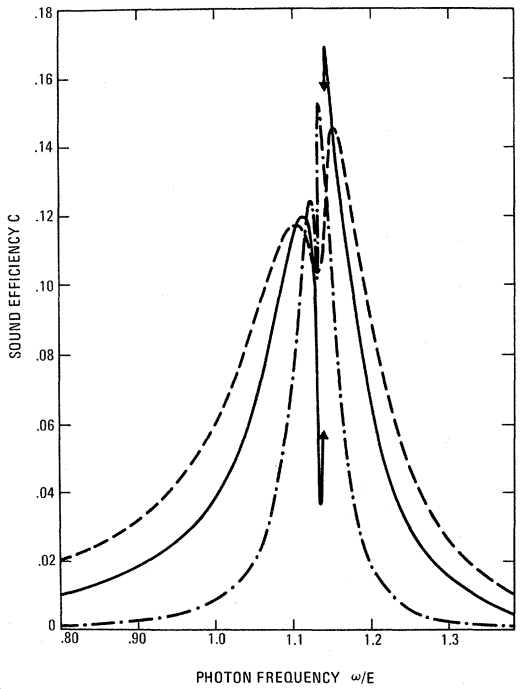

FIG. 8. Dependence of sound efficiency C vs ω/E on damping Γ/E for various values of parameters. Solid line: $A=0.5A_1$, $\Gamma/E=10^{-3}$; dashed line: $\Gamma/E=10^{-2}$; dash-dot line: $A=0.1A_1$, $\Gamma/E=10^{-3}$; all parameters not specified are the same as those in Fig. 1.

to include damping effects by incorporating appropriate damping terms in Eq. (20). Let us consider the effects of damping very generally without distinguishing between the different types[21] relating to exciton and phonon motion. For the latter case, damping may be included consistently[24] into the system equations of motion if ω is everywhere replaced by $\omega \to \omega + i\Gamma$, where Γ is an appropriate average damping constant (we neglect the real energy shifts of ω_0 and E which also arise). Carrying out this approximate procedure, which is valid for $\Gamma \ll \omega_0$, E, one obtains a rounding out of the peaks in $C(\omega)$, as would be expected in a damped system; typical results are illustrated in Fig. 8. The presence of large Γ could entirely wash out the double peak; however, the approximate theory described here is not reliable under such conditions, and a more rigorous calculation becomes necessary to obtain an accurate prediction. We emphasize also that the results for C illustrated in Fig. 8 refer to the efficiency at the exciting boundary of the crystal. At any point within the material the spatial damping of each wave with coefficient $\propto \mathrm{Im}[n_{1,2}(\omega)]$ need be incorporated to correctly obtain C. This may be done by straightforward generalization of certain well-known existing techniques.[25]

We have not discussed variations in v_s because, with $v_s/c \sim 10^6$ these have very little effect on the nature of the above results.

V. DISCUSSION

We have here presented a theory of excitation of sound by light which proceeds in the model adopted here via a combination of exciton-phonon and photon-exciton interactions such as may occur in narrow-gap semiconductors. Of special interest is the possibility of exciting a narrow band of high-frequency sound in a material by use of an appropriate laser source. The intensity as well as the frequency could be modulated directly by tuning the laser. The selective interchange of light energy to sound possible in such systems could conceivably be of eventual use in applications such as communications. Assuming the carrier density could be sufficiently diminished then in the particular case of the narrow-gap semiconductor varying the energy gap[26] by varying the pressure and temperature, say, could provide a method of varying the resonant frequency range of the semiconductor.

Our development indicates that the frequency dependence of C depends sensitively on the various microscopic coupling parameters characterizing the particular system under observation. A study of observed C values could thus provide information about the values of these parameters. Another way in which the measured values of C could be of use is in determining the nature of the microscopic boundary conditions appropriate for various processes. For example, "clamped" as opposed to "free" surface b.c.'s lead to certain qualitatively different behavior in C (see Fig. 7).

We summarize the various general properties revealed by the calculations which are expected to hold as well in other similar systems. In the absence of damping, if the derivative of the amplitude of the inactive field vanishes at the boundary surface, then the inactive-field fraction excited (F) peaks in the crossover frequency region. Correspondingly, the excitation efficiency (C) peaks doubly in the region, leaving a deep valley in the "reflection gap." The higher-energy peak dominates, except at low values of background dielectric constant. Higher photon-intermediate field and intermediate-inactive field couplings and lower background dielectric constant tend to broaden the resonance. The latter two variations substantially increase the maximum efficiency, while the first has a less pronounced effect. Increasing the damping from zero to finite values tends to eliminate the deep valley between the peaks in C; as damping increases the peaks round out more smoothly. All of the above features hold as well if the b.c. is changed to correspond to the inactive-field amplitude vanishing at the boundary, except that an additional low broad maximum appears on the lower-

energy side of the resonance in F and C.

For values of parameters appropriate to the specific problem at hand, the maximum in F or C is in the range of 10–20%. When damping is present one must also take into account the losses in amplitude as the excitations propagate through the material.[25]

We note an important relationship between C and R from an experimental point of view: The measured value of R provides information towards an experimental check or an actual determination of the value of p_1/p_2. (Of course, R alone does not determine p_1/p_2 uniquely except in special circumstances, such as when $\mathcal{E}_1/\mathcal{E}_2$ and the n_i are pure real.)

It is hoped that the present treatment will encourage experimental investigations of "indirect" excitation of quasiparticles by light, and that both the theoretical and applied aspects of such procedures can be further explored and exploited.

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*Supported by Office of Naval Research Grant No. NONR-N00014-69-A-0200-6206, NSF Grant No. GP-10943, and Air Force Office of Scientific Research Grant No. AFOSR-610-67.

†Present address.

[1]C. Kittel, *Quantum Theory of Solids* (Wiley, New York, 1963), Chaps. 2 and 3.

[2]M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Oxford U. P., Oxford, 1954), Chap. IV.

[3]Cf., e.g., M. Tinkham, *Group Theory and Quantum Mechanics* (McGraw-Hill, New York, 1964), Chaps. 7 and 8.

[4]A short but useful summary of helicon properties and references to more detailed works are given by J. J. Quinn and S. Rodriguez [Phys. Rev. 133, A1589 (1964)]; for a second-quantized treatment of helicons see P. Tannenbaum, Ph. D. thesis (New York University, 1969) (unpublished).

[5]J. J. Quinn, Phys. Rev. Letters 24, 817 (1970) and references therein.

[6]Transmission under oblique incidence would be necessary to directly excite plasmons. (a) See D. W. Berreman, Phys. Rev. 130, 2193 (1963); (b) M. Cardona, in *Semiconductors and Semimetals*, Vol. 3, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1967), p. 134. (c) Also, see A. Mooradian and A. L. McWhorter, in *Proceedings of the International Conference on Light Scattering Spectra of Solids*, edited by G. B. Wright (Springer-Verlag, New York, 1969); (d) A. L. McWhorter and P. N. Argyres, *ibid.*, and references in both these papers.

[7]C. Kittel, Ref. 1, Chap. 4.

[8]Cf., e.g., T. Moriya, J. Appl. Phys. 39, 1042 (1968).

[9]Y. Toyozowa, Progr. Theoret. Phys. (Kyoto) 20, 53 (1958).

[10]T. V. Ramakrishnan and L. J. Sham, in Proceedings of the International Conference on Narrow-Gap Semiconductors (unpublished).

[11]Cf. various papers on electronic properties of narrow-gap semiconductors in Ref. 10; also articles by C. Verie and L. Sosnowski and R. R. Galazka, in *II-VI Semiconducting Compounds*, edited by D. G. Thomas (Benjamin, New York, 1967) and references therein.

[12]Some early representative works are K. Huang, Nature 167, 779 (1951); U. Fano, Phys. Rev. 103, 1202 (1956); J. J. Hopfield, Phys. Rev. 112, 1555 (1958); L. N. Ovander, Usp. Fiz. Nauk 86, 3 (1965) [Sov. Phys. Usp. 8, 337 (1965)] and references therein to his and other earlier Russian work.

[13]For a discussion of a similar b.c. problem in relation to exciton excitation, cf., e.g., J. J. Hopfield and D. G. Thomas, Phys. Rev. 132, 563 (1963).

[14]R. Loudon, in Ref. 6(c).

[15]Calculations we have made indicate that as long as γ is a smoothly varying function of k, the exact form of the dispersion does not significantly influence the general nature of the results. When treating some particular material in detail, of course, it may be necessary to appropriately generalize or alter the above choices for the dispersions.

[16]In Ref. 2, such a division is made on the basis of physical arguments for a photon-phonon problem. Analogous arguments do not, however, present themselves in the present three-field case. Actually, we believe that the separation employed in Ref. 2, although a physically reasonable one, is inferior to that which would result in the present quantum-transformation method; we simply note that the two methods lead to results which are very similar qualitatively, differing only slightly quantitatively in the resonance regime.

[17]Cf., e.g., J. J. Hopfield in Ref. 12.

[18]Three places where such quantities are discussed in some detail are (a) D. L. Mills and E. Burstein, Phys. Rev. 188, 1465 (1969); (b) B. Bendow and J. L. Birman, Phys. Rev. B 1, 1678 (1970); and (c) B. Bendow, Ph. D. thesis (New York University, 1969) (unpublished). The latter discusses and computes various coefficients for a three-field system.

[19]Reference 18(b).

[20]See, e.g., A. Maradudin *et al.*, *Theory of Lattice Dynamics in the Harmonic Approximation* (Academic, New York, 1963), Chap. VI, and references therein to various papers by Wallis, and Wallis *et. al.*

[21]A. K. Ganguly and J. L. Birman, Phys. Rev. 162, 806 (1967).

[22]We may estimate the typical magnitudes of these constants employing (eV units) $P \sim \hbar/a$, $D \sim 10$, $\epsilon_b \sim 10$, $M/M_H \sim 10^2$, $a \sim 5$ Å, $a_0 \sim 30$ Å, and ω_0, $E \sim 10^{-2}$. These values lead to $\gamma' \sim 10^{-2}$–10^{-3} and $\beta/E^{1/2} \sim 10^{-1}$; i.e., the photon-exciton coupling may exceed the phonon-exciton by two orders of magnitude. However, we have used the upper-bound value for P, while typically this momentum matrix element can be very small in particular cases. Both γ and β are overestimated also in that we employ results for hydrogenic excitons; γ and $\beta \propto |\psi(0)|$, the relative-motion wave function of the electron-hole pair (Refs. 9 and 21). In the hydrogenic case, $|\psi(0)|^2 \sim 50$–75%, while this probability is clearly substantially less in highly screened systems such as the narrow-gap semiconductor. For example, if we assume that the screening reduces γ^2 and β^2 by an order of magnitude, and that P is an order of magnitude less than its maximum value, then for ω/ω_0, $\omega/E \sim 1$, $A \equiv \beta^2/E^3 \sim 10^{-1}$, while $B \equiv \gamma^{12}/E^2 \sim 10^{-2}$–$10^{-3}$. While these values should be kept in mind,

we will nevertheless vary parameters rather freely in order to most clearly illustrate various dependences. Evidently, the actual values of A and B can vary quite widely depending on the sizes of D, P, ϵ_b, $|\psi(0)|$, and M, for example.

[23]The analogy here is to a vibrating string with fixed vs free ends.

[24]This follows from an equation-of-motion analysis as detailed, e.g., by B. Bendow, Ref. 18(c).

[25]See, e.g., R. Loudon, J. Phys. 26, 667 (1965).

[26]General theory of gap variation due to pressure and temperature in semiconductors is discussed by E. J. Johnson, in Ref. 6(b), and references therein; for specifics regarding narrow-gap materials see Ref. 11.

PHYSICAL REVIEW B VOLUME 4, NUMBER 10 15 NOVEMBER 1971

Nonlocal Corrections to the Band Structure of Si, Ge, and α-Sn†

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(Received 29 June 1971)

The electronic structure of three group-IV elements is examined using a nonlocal pseudopotential determined primarily by physical considerations. An ionic contribution to the potential is separated from the valence and covalent portion of the interaction, and is constructed in an empty-core approximation, in terms of a set of angular-momentum–dependent core radii R_l. A modified version of the Penn dielectric function is used to determine the valence charge potential. Comparison with experiment results in substantial improvement over what has previously been achieved using the local empirical model (EPM).

I. INTRODUCTION

Over the past several years the pseudopotential method has proven to be effective in describing the electronic spectra of many covalently bonded semiconductors. In the early studies the Fourier coefficients of potential were treated as disposable parameters and fitted to selected experimental data.[1] The resulting analysis for a wide range of experiment was sufficiently good that one could be reasonably certain that no major revisions of the electronic structure of the materials studied would thereafter be necessary. Continuation of the original scheme to a variety of compounds by Cohen and collaborators[2] showed that the technique would work with the same effectiveness as for the group-IV materials. Further work by Saravia and the author[3] demonstrated that one could in addition successfully calculate deformations of the electronic structure associated with hydrostatic and uniaxial strains.

Several considerations make it worthwhile to reexamine the electronic structure of the group-IV materials. Perhaps the most compelling of these is the central role played by these substances in the Periodic Table. For example, given the electronic structure of Ge, one would hope to be able to get those of GaAs, ZnSe, CuBr, etc., as a series of well-defined ever-increasing perturbations on that of the central member of the sequence. Indeed, within a pseudopotential framework such a start has already been made by the author.[4] Using a nearly self-consistent model, he found it possible to construct potentials for several of the III-V compounds without the use of any empirical inputs other than those provided by the group-IV elements. Comparison was made between calculated and experimental values for 42 electronic transitions in these materials. The over-all agreement was as good as that achieved through either first-principles[5] or purely empirical methods.[6] It is natural, therefore, to suppose that any improvements in the pseudopotentials for the group-IV crystals will put us in a position to do more definitive work in the related AB compounds.

Examination of the current situation in Ge will suffice to indicate the present state of the art regarding band-structure theory in semiconductors. The original local version of the pseudopotential was able to specify the important energy levels to an accuracy of about 0.1 eV.[1] There were, however, a few levels that could only be located to within something slightly better than 0.5 eV. Of most concern at the time was the large optical-reflectivity peak near 4.3 eV. This appeared to be 0.4 eV too low in the theoretical spectrum.[1,7] A subsequent paper using the pseudopotential method did not meet with any more success in improving upon this problem.[2] Passing on to electronic multiplet associated with the $\Gamma_{25'} \rightarrow \Gamma_{15}$ transition, we note that considerable and intensive efforts have been made recently which apparently finally resolve this elusive transition. Donovan *et al.*[8] have made an exhaustive examination of the leading edge of their photoemission EDC's and place the center of mass of the multiplet slightly below 3.2 eV. The same authors, noting the lack of polarization dependence of their transverse electroreflectance signal, definitively assign the 3.2-eV multiplet to this Γ excitation. Careful piezo-optic measurements also place the gap at the above