Phys. Rev. B 4, 4417 (1971).
[43]T. M. Donovan, Ph.D. thesis (Stanford University, 1970); Stanford Electronics Laboratories Report No. SU-SEL-70-047, 1970 (unpublished); T. M. Donovan and W. E. Spicer, Phys. Rev. Letters 21, 1572 (1968); W. E. Spicer and T. M. Donovan, J. Non-Cryst. Solids 2, 66 (1970).
[44]D. T. Pierce and T. H. DiStefano, Rev. Sci. Instr. 41, 1740 (1970).
[45]J. Matsuzaki, Stanford Electronics Laboratories Report No. SU-SEL-70-084, 1970 (unpublished).
[46]R. J. Powell, J. Appl. Phys. 41, 2424 (1970).
[47]F. Wooten (private communication).

PHYSICAL REVIEW B VOLUME 6, NUMBER 4 15 AUGUST 1972

Coulomb Interaction in Semiconductor Lasers

W. D. Johnston, Jr.
Bell Telephone Laboratories, Holmdel, New Jersey 07733
(Received 21 April 1972)

Recent evidence suggests that the excitation density in semiconductor lasers exceeds the realm where exciton-based descriptions of the optical-gain process are valid. A treatment is presented based on the random-phase approximation from electron-gas theory in which the effects of the long-wavelength components of the Coulomb potential are included. In addition to the usual exchange term, a term derivable from electron-plasmon coupling modifies the electron self-energy (and gives an apparent "band-gap shift") in the range of density parameter $1 \leq r_s \leq 5$ which appears to characterize semiconductor lasers. Agreement of prediction with experimental wavelength shifts for stimulated emission in an external magnetic field for GaAs and $CdSnP_2$ is excellent. The average electron self-energy shift from the long-wavelength part of the Coulomb interaction may be simply interpreted as arising from the plasma zero-point energy.

I. INTRODUCTION

A multitude of reports in the literature now confirm that virtually all direct-band-gap–semiconductor compounds of the III-V, II-VI, and even I-VII types exhibit signs of stimulated emission (ranging from strong superradiance to unimpeachable laser action) under appropriate excitation. Nevertheless, the mechanism of the optical-gain process remains obscure. Particularly in the case of the II-VI compounds, but in other materials as well, numerous models based on a variety of exciton recombination possibilities have been proposed. While each of these suggestions appeared to account for some set of data, neither singly nor in combination have they been able to account for the ever-growing total body of experimental observations. One of the simplest features of a useful model would appear to be the prediction of the wavelength position and width of the optical-gain region. Ideally, of course, one would like to be able to give exactly the optical-gain spectrum as a function of temperature, excitation, etc.

There are common features to the stimulated emission from diverse (direct band-gap) materials which suggest that particularly material-dependent or extrinsic properties are not of fundamental importance. This is further borne out by the observation that the stimulated emission is not ordinarily confined to a narrow spectral region (narrow, for example, in the sense of being comparable to a spontaneous exciton emission line), nor is the emission fixed at a constant difference from the band gap as excitation or temperature is varied. While isolated exceptions may exist, we will consider here that the optical-gain process in excited semiconductors with direct band gap is generally an intrinsic phenomenon. We have thus to consider direct, or band-to-band, carrier recombination plus intrinsic exciton recombination processes.

Of the latter, exciton-phonon,[1] exciton-electron,[2,3] exciton-exciton,[2,3] and excitonic-molecule[4] processes have been proposed, with varying degrees of success, to account for observed stimulated emission behavior. These models are theoretically valid only in the low-excitation-density regime, where "hydrogenlike" electron-hole bound states exist. Recent developments, both theoretical and experimental, converge to cast considerable doubt that exciton-based models are valid for the semiconductor laser excitation regime (see Sec. II).

The alternative many-body approaches to electron-hole interactive recombination, suggested initially by Basov *et al.*,[5] have received much support from the experimental observations[6] of Holonyak and his co-workers, who conclude that the stimulated emission in GaAs, CdSe, and other materials cannot be explained by exciton processes, impurities, or sample heating. Recently, the first

evidence of a clear quantum-plasma effect associated with spontaneous and stimuated emission was presented.[7]

In this paper we wish to present a theory derived from the random-phase approximation appropriate for the excitation-density regime of semiconductor lasers. Electron-plasmon coupling and exchange interaction are found to be important effects. Comparison is made with experimental results for $CdSnP_2$ ($n \sim 10^{17}$ cm^3)[7,8] and high-purity GaAs,[9] and the theoretical development underlying the interpretation of Ref. 7 is amplified.

Finally, the difficulties in making contact between the experimental results and the theory (problems of geometry, nonuniform pumping, high photon density and saturation) are discussed. These difficulties are generally nontrivial and apparently still not widely appreciated.

II. EXCITATION DENSITY AND EXCITON STABILITY IN SEMICONDUCTOR LASERS

The volumetric rate of electron-hole pair creation in an excited semiconductor can ordinarily be estimated accurately from absorption data and a knowledge of the absolute intensity distribution of the exciting source. This is particularly true when optical excitation by a "well-behaved" pump laser with a simple, or at least reasonably uniform, spatial mode distribution is employed. There is however, a problem in estimating the steady-state electron-hole density, since the lifetime of an excitation, under the particular stimulated emission condition of the experiment, cannot be predicted from elementary considerations and should be measured. Such measurements as have been made directly[10] show typical lifetimes in the stimulated regime of the order of tenths of nanoseconds, and imply threshold excitation densities from 10^{17} well into the 10^{18}-cm^{-3} range, for III-V and the narrower-gap II-VI compounds. Less direct information[11,12] implies similar numbers for the wider-gap II-VI compounds as well.

Excitation densities of this order or greater present clear problems for any exciton-based description of the optical-gain mechanism. An analysis published recently by Gay[13] shows that in theory the hydrogenic-exciton state ceases to exist at a lower carrier density than had heretofore been estimated. In fact, for CdS he shows that the free-exciton binding energy vanishes at an electron concentration of $\sim 10^{17}$ cm^3. In terms of the electron-density parameter characterizing the high-density plasma,

$$r_s \equiv (\tfrac{4}{3}\pi a_0^3 n)^{-1/3} \quad , \tag{1.1}$$

where $a_0 = (\epsilon_0 \hbar^2/e^2)(1/m_e^* + 1/m_h^*)$ is the excitonic Bohr radius and n the carrier density, Gay's calculation corresponds to a critical value of $r_s = 5$ for CdS. By way of comparison, the Mott transition[14] occurs at $n_{\text{Mott}} = (64\pi a_0^3)^{-1}$ or $r_s \approx 2.5$ for a single carrier gas, and at $r_s \approx 10$ for an equal mass isotropic electron-hole plasma.[15] This latter value is ordinarily taken as an order-of-magnitude estimate of the donor concentration at which donor levels merge to form a conduction-band tail. Since r_s provides the proper scaling for screening and other many-body effects in the high-density regime, we expect that exciton models will be useful only at excitation densities such that $r_s > 5$, not only for CdS, but for semiconductors generally. This should be true for nonhydrogenic excitons (such as in CuCl) as well if a_0 is taken as the experimental value for the $n = 1$ exciton level. (The levels of the finite-range screened-Coulomb potential are in any case "nonhydrogenic.")

In Table I is summarized the excitation density, as calculated from measured or estimated effective excitation lifetimes, corresponding to threshold for laser action, to onset of superradiant emission, or to other indications of significant stimulated emission, for a variety of representative semiconductors. It is interesting to note that in every case the deduced values for r_s lie below even the value for Mott transition. The weak dependence of r_s on n suggests that even though there is uncertainty for the excitation density in some materials, the conclusion that lasing takes place in a non-excitonic regime remains valid. From another point of view, the similarity of the values of r_s at threshold in Table I are consistent with an approach to semiconductor optical gain from a universal (i.e., not material specific), electron-gas theoretical point of view.

III. OPTICAL GAIN AT HIGH EXCITATION DENSITY, LIMIT $r_s \ll 1$

Before discussing the region of actual interest, $1 \lesssim r_s \lesssim 5$, it is useful to review the nature of the optical gain to be expected in the high-density limit, in which the Fermi energy dominates either thermal or Coulomb energy: $E_F \gg T$, $e^2 p_F$. [In this and subsequent sections, $\hbar = 1$, e^2 is the square of electronic charge reduced by lattice dielectric constant, Boltzmann's constant is 1, $p_F \equiv (3\pi^2 n)^{1/3}$ is the Fermi momentum, and E_F is the noninteracting electron gas Fermi energy.] We may use the ground-state Green's-function formalism in this case.

The relevant calculation has been made by Gergel.[16] He writes

$$\chi(\omega, \vec{k}) = -i|d|^2 \int \frac{d^4p}{(2\pi)^4} G^1(p + \tfrac{1}{2}k) G^2(p - \tfrac{1}{2}k) \, , \tag{3.1}$$

with χ the optical susceptibility at frequency ω and wavevector $\vec{k}$, d the dipole interband-transition

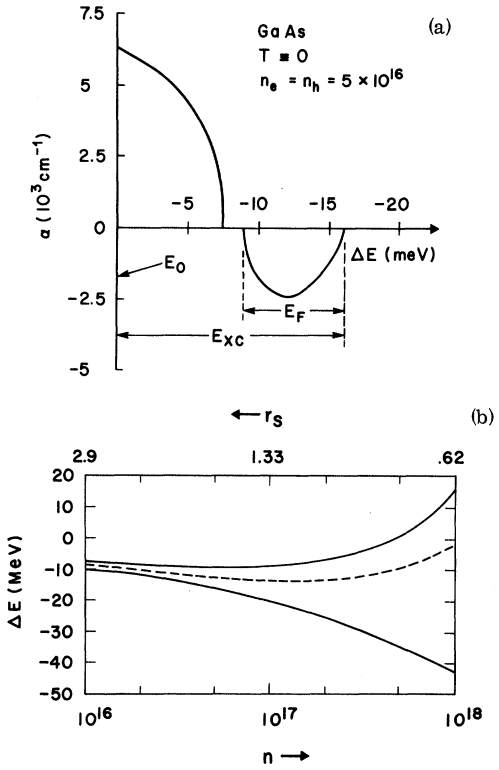

FIG. 1. (a) Optical-absorption coefficient α in approximation of intrinsic GaAs; $n_e = n_h = 5 \times 10^{16}$, $|d|^2$ computed from Ref. 15, $T = 0$. (b) Region of gain in intrinsic GaAs, $m_e/m_h \sim 10$, as in Ref. 13, $T = 0$. The dashed line indicates peak gain position for photon $k = 3.5 \times 10^5$ cm^{-1}.

matrix element, and $G^{1,2}$ the single-particle electron (hole) Green's function including correction for the electron-hole pairing instability[17] arising from the screened Coulomb potential.

The matrix element $|d|$ may be obtained, for purposes of absolute calculation, from the electron and hole effective-mass values.[18] Optical gain results, of course, in the region where $\mathrm{Im}\chi < 0$. Equation (3.1) may be solved analytically if the photon dispersion is ignored, $k = (k^4, \vec{k}) = (\omega, 0)$; or for $\vec{k} \neq 0$ if electron and hole effective masses are equal. Otherwise numerical solution is possible.

A typical result is sketched in Fig. 1, using parameters appropriate for GaAs for constant finite k (i.e., the change in k due to $\mathrm{Re}\chi$ is ignored). A gain region exists which may lie below, or partly below and partly above, the dilute or unexcited band gap E_0. The lower edge is at $E_0 - 4e^2 P_F/\pi$, and drops as $n^{1/3}$. The upper edge is given by the Fermi level to be at $E_0 - 4e^2 p_F/\pi + \frac{1}{2} p_F^2 (1/m_e^* + 1/m^*)$. The particular feature introduced by the screened Coulomb electron-hole interaction is a "gap" within which χ vanishes between the gain and absorptive regions, the width of which gap vanishes exponentially as n increases. This gap is small and is expected to "wash out" at finite temperature (in fact at ~ 1 °K for GaAs at 10^{17} cm$^{-3} = n$). Electron-electron and/or hole-hole vertex corrections were shown to be less important in Ref. 17, so that only the exchange interaction would appear important.

In spite of its simplicity, Gergel's description has qualitatively most of the features needed to predict the experimental wavelength results. Also, a maximum gain of $\sim 10^4$–10^5 cm^{-1} is possible, since matrix elements for scattering with phonons or other carriers are not directly involved. A relatively broadband gain region emerges, which increases in width with increasing excitation. In Gergel's treatment the long-wavelength edge moves always to the red with increasing n, with the upper edge moving first to the red and then to the blue at sufficiently large values of E_F, in accord with the usual red shift (and exceptional blue shift in strongly n-type material) observed experimentally as temperature or excitation is increased. This motion of the gain region as a function of n is shown in Fig. 1(b), again with material values for GaAs.

There are, of course, shortcomings of this model in detail. The wavelength shifts are not quantitatively as well in accord with experiment as one would like. The "high-density limit" is operative first in the restriction of the model to $T = 0$, but second, in the form of interaction potential used in Ref. 17, where the shielded, rather than the full Coulomb potential, was employed. Thus effects (other than the shielding) due to the low-momentum-transfer components of the Coulomb interaction have been ignored. This is well-known to be proper in the high-density limit, but (as we show in Sec. IV) an important self-energy correction to the electrons arises therefrom in the $1 \lesssim r_s \lesssim 5$ range. The fact that the only other important self-energy correction (i.e., the exchange energy) is included in Gergel's treatment accounts for its success, particularly as this is the only red-shifting term which arises from a many-body calculation.

IV. SELF-ENERGY CORRECTIONS AND OPTICAL GAIN AT INTERMEDIATE DENSITY, $1 \leq r_s \leq 5$ AND $T \neq 0$

In this section a calculation relaxing the high-density restriction of Gergel's presentation is developed. The optical gain arises as in Ref. 19,

$$\mathrm{Im}\chi(\vec{k}, \omega; T, \mu) = |d|^2 \mathrm{Im}G_2^R(\vec{k}, \omega; T, \mu) = |d|^2 \tanh(\omega/2T)\, \mathrm{Im}G_2(k, \omega; T, \mu), \quad (4.1)$$

where G_2^R and G_2 are the Fourier transforms of the "retarded" and "causal" two-particle Green's

functions, respectively:

$$G_2^R(k,\ \omega) = \int d^3\vec{r}\, dt\, e^{i(\omega-\mu_e+\mu_h)t - i\vec{k}\cdot\vec{r}}\, G_2^R(\vec{r},\ t)$$

$$G_2^R(r,\ t) = \begin{cases} \langle T \,|\, \Psi_h^\dagger(\vec{r},\ t)\,\Psi_e(\vec{r},\ t),\ \Psi_e^\dagger(0,\ 0)\,\Psi_h(0,\ 0)\,|\rangle, & t>0 \\ 0, \quad t<0 \end{cases}$$

$$G_2(r,\ t) \quad -\langle T \,|\, \Psi_h^\dagger(\vec{r},\ t)\,\Psi_e(\vec{r},\ t),\ \Psi_e^\dagger(0,\ 0)\,\Psi_h(0,\ 0)\,|\rangle \quad \text{all } t.$$

As usual, $\langle T\,|\cdots|\,\rangle$ stands for a statistical average of the time-ordered quantity therein. The last relation in (4.1) follows from the fact that the Fourier transform of a two-fermion Green's function with coincident arguments, i.e., $G(x_1x_2;\ x_3 = x_1,\ x_4 = x_2)$ has the analytic behavior of a boson propagator.[20]

We may now write[21]

$$G_2(k) = (2\pi)^{-4}\int G_1^{e1}(p+\tfrac{1}{2}k)\,G_1^h(p-\tfrac{1}{2}k)\,d^4p + \text{vertex terms.} \quad (4.2)$$

The "vertex terms" were discussed in Ref. 17. They correspond to higher-order scattering or propagation of an excitonlike electron-hole bound state. The energy of this state corresponds to the gap discussed in Sec. III, and we expect it to be unimportant at finite temperature. We write simply

$$G_1(\vec{k},\ \omega) = [\omega - \omega(\vec{k}) - \mu_{e1} + \mu_h - \Sigma(\vec{k},\ \omega)]^{-1}, \quad (4.3)$$

where $\omega(k) = \pm(k^2 - k_F^2)/2m_{e,h}^*$ for electrons or holes, $\mu_{e1} - \mu_h = E_0 + \mu(T, N_{e1}) - \mu(T, N_h)$ gives the forbidden width between quasi-Fermi levels of electrons in the valence band and holes in the conduction band.

We now consider the self-energy corrections $\Sigma(k)$. The details depend on whether the electrons and holes are both degenerate, or not, and whether the effective masses are comparable or not. We will use the random-phase approximation for the self-energy shown in Fig. 2. The "rings" may now be either electron or hole (valence-band) rings, in general. We have as usual

$$\Sigma(p) = \int \frac{d^4q}{(2\pi)^4}\ \frac{V(q)G^0(p-q)}{1 - V(q)\,\Pi(q)}\ ,$$

with $\Pi(q) = \Pi_{e1}(q) + \Pi_h(q)$, $V(q) = 4\pi e^2/q^2$, and

$$\Pi_{e1,h}(q) \equiv 2\int \frac{d^4p}{(2\pi)^4}\ G^0_{e1,h}(p)\,G^0_{e1,h}(p+q)\ .$$

The G^0 are the noninteracting electron and hole Green's functions.[21]

For sufficiently n-type material we may ignore the presence of holes in the valence band, and carry over the well-known results for Σ in an electron gas. Detailed calculations have been made by Lundqvist,[22] particularly emphasizing the effects

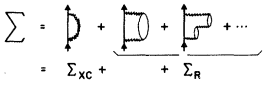

FIG. 2. Proper electron or hole self-energy contributions in random-phase approximation—exchange term (Σ_{xc}) plus electron and/or hole ring terms (Σ_R).

from the small-momentum-transfer region of $V(q)$ in the range of $1 \lesssim r_s \lesssim 5$ in which we are interested. We consider this case first, and then note the modifications required for high-purity material where the plasma may have two important components.

It is useful to break the self-energy into three types of terms:

$$\Sigma(p) = \Sigma^0 + \Sigma(|\vec{p}|^2) + \Sigma'(p,\ \epsilon)\ . \quad (4.4)$$

By comparison with Gergel's results we see that the first term independent of $\vec{p}$, p^4 will rigidly shift the gain region. Such a term comes from the first (Σ_{xc}) diagram of Fig. 2. The second $\sim(p)^2$ term simply modifies the effective mass, while the other terms contribute interesting structure representing new elementary excitations such as the plasmaron, etc. It is also useful to consider the shift in thermodynamic potential $\delta\Omega$, which may be thought of as representing an "average" self-energy correction

$$n\,\langle\Sigma(p)\rangle \equiv \delta\Omega(T,\ \mu) = \int \frac{d^4p}{(2\pi)^4}\,\Sigma(p)G^0(p)\ , \quad (4.5)$$

and to write

$$\Sigma(p) = \Sigma_{xc} + \langle\Sigma_R\rangle + \Sigma'(|\vec{p}|^2) + \Sigma''(p,\ \epsilon)\ . \quad (4.6)$$

Thus, if we are only interested in *shifts* of the gain region, the first two terms of (4.6) suffice; the others simply alter the *shape* of the gain curve.

There are two reasons why the sharp features in $\Sigma(p)$ are not of particular concern to us here. These features are "washed out" in (4.2) for finite photon wave vector k, just as with the singularity at the Fermi surface in Gergel's treatment. Second, the experimental situation does not presently permit unambiguous confirmation of features in the gain-curve shape, as will be discussed in Sec. VI. The experimental shifts of stimulated light output, however, whether defined in terms of center of gravity of laser modes, or peak of superradiance, or whatever, should shift in accordance with $\langle\Sigma(p)\rangle$. This is a particularly convenient quantity to compute, for there is a simple representation in terms of the dielectric function of the medium.[23] Strictly (4.5) should be written as an integral over a variable coupling parameter, λ; then

$$\Omega = \int_0^\lambda d\lambda \int \frac{d^4p}{(2\pi)^4}\,\Sigma^\lambda(p)\,G^0(p)$$

$$= \int d\lambda \int \frac{d^4p}{(2\pi)^4} \int \frac{d^4q}{(2\pi)^4} \frac{V^\lambda(q)}{1 - V(q)^\lambda \Pi(q)} \times G_0(p)\, G_0(p-q), \quad V^\lambda(q) = \lambda V(q)$$

$$= \int_0^1 d\lambda \int \frac{d^4q}{(2\pi)^4} \frac{V^\lambda(q)\,(q)}{1 - V^\lambda(q)\,\Pi(q)}$$

$$= \int_0^1 d\lambda \int \frac{d^4q}{(2\pi)^4} \left(\frac{1}{\epsilon^\lambda(q)} - 1 \right) , \tag{4.7}$$

where $\epsilon^\lambda(q) \equiv 1 - V^\lambda(q)\,\Pi(q)$ is the "ring-diagram" dielectric constant. The contribution from the exchange diagram in Fig. 2 may be separated explicitly:

$$\Omega = \Omega_{xc} + \Omega_R$$

$$= \int_0^1 d\lambda \int \frac{d^4q}{(2\pi)^4} V^\lambda(q)\, \pi(q) + \int_0^1 d\lambda \int \frac{d^4q}{(2\pi)^4} \frac{[V^\lambda(q)]^2 \pi^2(q)}{1 - V(q)\pi(q)}$$

$$= -4\pi e^2 \iint \frac{d^3\vec{p}\, d^3\vec{q}\, n_0(\vec{p})\, n_0(\vec{q})}{(2\pi)^6 |\vec{p} - \vec{q}|^2} + \Omega_R \ .$$

In the limit $T \to 0$, the first term gives a modified Hartree-Fock exchange energy $(2e^2/\pi)(p_F^{el} + p_F^h)$, just as in Ref. 16. The second term may be written as[22]

$$\Omega_R = \int_0^1 d\lambda \int \frac{d^4q}{(2\pi)^4} \left[\mathrm{Im}\left(\frac{1}{\epsilon^\lambda(q)} \right) + \mathrm{Im}\epsilon^\lambda(q) \right] . \tag{4.8}$$

The long-wavelength contribution from the Coulomb interaction arises from the plasma pole (poles, if the plasma is multicomponent or a magnetic field is present) of $1/\epsilon$. In terms of (4.3), these come from the high-order ring diagrams; the "bubble-chain" superposition represents a plasmon in this sense. This aspect may be emphasized by writing (4.8) in the alternative form[22]

$$\Omega_R = \int_0^1 d\lambda \int \frac{d^4q}{(2\pi)^4} G^0(q) \times \left(\int \frac{d^4k}{(2\pi)^4} |g_k^\lambda|^2 \frac{2\omega_k^\lambda}{\omega^2 - \omega_k^{\lambda 2} + i\delta} G^0(q-k) \right) , \tag{4.9}$$

where $2\omega_k/(\omega^2 - \omega_k^2 + i\delta) \equiv D(k)$, the plasmon propagator, and $|g_k^\lambda|^2 \equiv V^\lambda(k)[\partial\epsilon^\lambda(k, \omega_k)/\partial\omega]^{-1}$, the "plasmaron" coupling, have been introduced. The specific contribution in (4.9) may be regarded as deriving from an *approximate* effective Hamiltonian

$$H_{eff} = \sum_k \epsilon(h)\, C_k^\dagger C_k + \sum_q \hbar\omega_q (b_q^\dagger b_q + \tfrac{1}{2}) + \sum_{kq} (Vol)^{-1/2}\, g_q\, C_{k+q}^\dagger C_k (b_q + b_{-q}^\dagger)$$

$$= H_{el}^0 + H_{plasmon}^0 + H_{el\text{-}plasmon} \tag{4.10}$$

representing fermions coupled to a boson field. This is the same effective Hamiltonian form as that for electron–LO-phonon (polaron) coupling. An important difference from the polaron case is that the plasmon modes are not (as the lattice phonons are) independent of the quasielectron modes, since *all* of the physical electrons contribute to *each* plasma mode and vice versa. Thus, for instance, in the polaron problem, the zero-point energy of the lattice ($\lim_{T\to 0} \langle H_{phonon}^0 \rangle$) does not affect the electrons, while in the plasmaron problem the term $\langle H_{plasmon}^0 \rangle$ obviously does. Expressed differently, $H_{plasmon}^0$ and $H_{el\text{-}plasmon}$ are *both* proportional to the Coulomb coupling-strength parameter λ. The contribution from (4.9) is patently a result of electron-plasmon (plasmaron) coupling, corresponding to the self-energy diagram of Fig. 3, regardless of the specific interpretation placed on the separate terms of the approximation (4.10).[24]

The structure of this self-energy term from the long-wavelength ring-diagram contribution [large-bracketed portion of (4.9)] has been examined in ground-state formalism by Lundqvist in detail.[22] A numerical analysis based on his results confirms our earlier statement that self-energy "structure" is washed out in the integration in (4.2), essentially because the poles of the two single-particle Green's functions are noncoincident at finite photon momentum except for isolated values of the integration variable. In particular, the discontinuities and logarithmic singularities in the self-energy at $\omega = \omega(p) \pm \omega_q$ for $p \gtrless p_F$, corresponding to propagation of the plasmaron state,[19] do not give rise to sharp structure even in the ground-state calculation. Finite temperature can obviously cause only further smoothing.

Horing has shown[23] that in the simplest approximation $\Sigma(k) \to -(4\pi me^2/m^*\omega^2)\,\theta(k_c - k)$, Eq. (4.8) yields a value for $\langle \Sigma(p) \rangle$ of $(\pi me^2/m^*)^{1/2}(k_c/k_F)^3$. In general, it is straightforward to show from (4.8) or (4.9) that the contribution to $\langle \Sigma(p) \rangle$ is just

$$\langle \Sigma(p) \rangle_R = (1/2n) \sum_{q<q_c} (\omega_q) \ .$$

The value of the "cut-off" momentum q_c is determined by the onset of strong damping or other effect which invalidates the particular approximation used for the dielectric function. A precise (within the random-phase approximation, that is) expression for ϵ^{-1} has been derived by Horing[25] including ar-

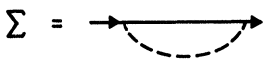

FIG. 3. Self-energy contribution from electron-plasmon (dashed line) coupling in lowest order corresponding to large parentheses in Eq. (4.9).

bitrarily strong external magnetic field and finite temperature.

Inclusion of magnetic field effects provides a convenient way to seek experimental verification of this model. From Mermin and Canel's work,[26] for instance, we know the pole arising from the upper hybrid plasmon will make most of the contribution to (4.8), and the momentum sum will emphasize plasmons propagating perpendicularly rather than parallel to any magnetic field. Thus, a contribution is expected $\langle\Sigma(p)\rangle \sim (\omega_p^2+\omega_c^2)^{1/2}K$, where K is a numerical constant of the order of one-half, ω_p and ω_c are the plasma and cyclotron frequencies, respectively. Any particular case of interest may be computed numerically, of course. It appears that K does not vary by more than a factor of 2 for values of r_s between 1 and 5, or for any relative values of ω_c to ω_p, including the effects of plasmon dispersion and/or Landau damping. In the region $\omega_c \sim \omega_p$, there is a "dilution" of the gain owing to the importance of the anisotropy term in the expression for the plasmon dispersion, but no other interesting effects are associated with the coincidence of ω_p and ω_c.

Let us summarize the results of this section. We have outlined how the well-known results from treatments of the interacting electron-gas problem can be applied to the determination of optical gain in semiconductors. A useful simplification is the replacement of the electron self-energy by the correlation energy in the corrected single-particle propagators. An important term from the long-range interaction corresponds to the zero-point plasmon energy, which is equal to $\frac{1}{2}\hbar\omega_p$ times a numerical factor about equal to unity. The precise determination of that factor is limited by the limitations of the random-phase approximation, which is strictly valid only for $r_s<1$ and for momenta $k<0.47r_s^{1/2}k_F$. Our treatment assumes extension into the "metallic" regime $1<r_s<5$, $k\sim k_F$, which is commonly made but requires interpolative procedures.[27]

We have ignored short-range electron-hole interaction, since there are obviously no exchange effects and since the energy gap from short-range electron-hole correlation is small relative to thermal energies for $r_s<5$. The possibility of electron-hole "drop" formation or plasma condensation[28] has also been ignored, owing to the expected low value of critical temperature for this phenomenon. The experimental observation that stimulated emission wavelength changes with excitation intensity argues against formation of a constant-density condensed phase in the semiconductor laser regime. Nevertheless, it is clear that as r_s increases above unity, the short-range electron-hole interaction eventually must dominate and yield exciton behavior. The approximation (4.2) for the two-particle Green's function will then clearly fail. A smooth theoretical connection of the "exciton" to the "plasma" behavior in the electron-gas–hole-gas mixture has not yet been realized and appears to be inherently incompatible with a perturbation-theoretical treatment.

V. COMPARISON WITH EXPERIMENT

Strictly speaking, since we have calculated what corresponds to a small signal optical-gain spectrum, comparison with experiment would be significant only if saturation effects were negligible. This is obviously not the case for semiconductor laser oscillators, and does not generally seem reasonable for the superradiant regime either. Nevertheless, stimulated emission must clearly come from somewhere within the small signal gain characteristic. We assert that at least for suitably small variation of the plasma-pole frequency, the gain region and stimulated *output*, regardless of saturating effects, will track; i.e., we assume the saturating mechanism is independent of small changes in exchange or correlation energy. We may vary the plasma-pole frequency by varying the density of equilibrium or nonequilibrium carriers, or varying an external magnetic field. The former approach is not particularly clean since there is not necessarily any simple relation between excitation flux and excitation density, and since exchange as well as correlation terms are modified. Dopant changes modify not only the majority carrier concentration but also the nonradiative recombination efficiency, nonresonant absorption, etc., as well as introducing impurity shifts of the band gap.

A more convincing demonstration may be obtained from the magnetic-field-shift data shown in Fig. 4. We note that the exchange-only theory would predict a blue shift proportional to the cyclotron frequency only. Exciton-exciton[3] processes would shift to the red with increasing magnetic field, and are absolutely excluded by the data for both $CdSnP_2$ and high-purity GaAs. For GaAs the electron density was determined from the exciting flux, but there is enough data in fact to permit self-consistent determination of the effective carrier density. With optical excitation the density is almost certainly nonuniform, and probably varies rapidly enough that the optical propagation characteristics of the pumped semiconductor are affected,[29] so that an "effective" density of carriers must be used in any case. In the GaAs case the plasma frequency is taken as $[4\pi ne^2(1/m_e^*+1/m_h^*)]^{1/2}$ since the number of electrons and holes are equal.

For both $CdSnP_2$ and GaAs the magnetic-field-shift data are in excellent agreement with the prediction of a shift proportional to $(\omega_p^2+\omega_c^2)^{1/2}$. In both materials the characteristics of the radiation are similar in that the peak wavelength shifts with

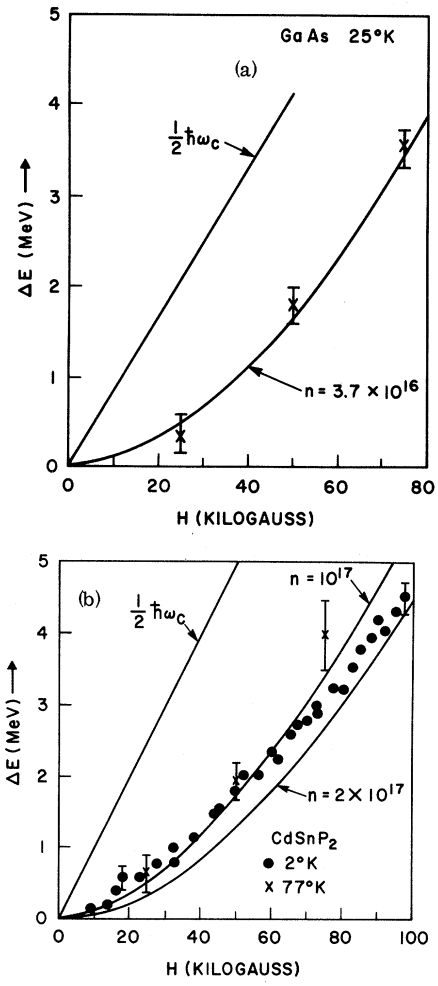

FIG. 4. Wavelength shift of stimulated emission for (a) high-purity GaAs and (b) n-type $CdSnP_2$ in magnetic field. Solid curves for $\langle\frac{1}{2}\hbar\omega_+\rangle$. For GaAs ($T=25$ °K), $n_e=n_h=3.7\times10^{16}$, $\epsilon=12.9$, $m^*=0.07m_0$. For $CdSnP_2$, $m^*=0.06m_0$, $\epsilon=12$, $n_h\ll n_e=1.5\times10^{17}$ ($T=2$ °K, ●; $T=80$ °K, ×). Data from Refs. 7–9.

increased excitation to the red, and the width of superradiance increases. In GaAs "quenching" of the superradiance occurs sooner (at ~45 kG), as one would expect when $\omega_p\sim\omega_c$. It is interesting to note that collisional damping effects should be much more important in $CdSnP_2$ where $\omega_p\tau\sim1$ (with τ a collision time from mobility data) than in the high-purity GaAs where $\omega_p\tau\sim10^2$. We do not see any indications suggesting that collisional damping of the plasmon or cyclotron motion is important. We conclude that the stimulated recombination in $CdSnP_2$ and high-purity GaAs (also *a fortiori*, n-type GaAs) is band-to-band, direct recombination with Coulomb interaction giving rise to "band-gap shifts" corresponding to the exchange and long-range correlation energies of a dilute plasma. (The number of particles per Fermi-Thomas sphere is $0.735r_s^{-1/2}$, $\lesssim1$ for $1\lesssim r_s\lesssim5$.)

We recall that other workers have observed initially quadratic blue shifts of laser output with magnetic field InSb,[30] InAs,[31] and GaAs diodes.[32] For small values of ω_c/ω_p, the expression $(\omega_p^2+\omega_c^2)^{1/2}$ shifts quadratically with magnetic field becoming linear at high fields. The results for InSb[30] and InAs[31] are qualitatively of this sort. The GaAs results[32] did not show a linear region and were interpreted in terms of a possible diamagnetic shift of donor levels. Determination of the effective majority carrier concentration in the active region of a laser diode is extremely difficult, and is further complicated by the effect of magnetic field on junction current. We wish only to point out that these early observations on diodes are at least qualitatively in accord with the many-body treatment described here. We will not attempt to establish the quantitative accord which appears possible.

The temperature dependence of the gain from this model is unfortunately not simply expressible. It is of the form

$$\mathrm{Im}\chi_{\max}\sim\tanh^{-1}(\mu/kT)F(T)\,,$$

with $\mu(T)$ the quasi-Fermi level of the majority carrier. $F(T)$ is proportional to the inversion. If electrons and holes are equally degenerate ($m_e=m_h$), $F(T)$ is roughly proportional to the number of electronic states with occupation probability greater than one-half; i.e., to

$$\int_0^{\mu(T)}[n_\mu(E)-\tfrac{1}{2}]g(E)\,dE\,,$$

with $n_\mu(E)$ the Fermi-distribution function and $g(E)$ the density of states. The temperature dependence of an experimentally determined threshold will depend on the cavity losses, and can be quite different even for the same sample pumped optically or by electron beam, since the temperature dependence of the gain and the *level* of the gain are interrelated. (Optical pumping generally requires considerably higher excitation for the same *net* gain due to diffraction effects, as shown in Ref. 29.) This is in contrast to the temperature dependence for exciton-based gain mechanisms which is invariant in functional form, assuming the excitons are described by Boltzmann statistics, regardless of excitation density. Thus a "characteristic temperature" for exciton-based gain processes is a constant, while for band-to-band recombination the "characteristic temperature" scales with $n^{2/3}$. Thus one will have, in general, a different apparent temperature dependence for threshold in a "high-threshold" than in a "low-threshold" configuration, even for temperature-independent nonresonant loss.

Experimental threshold measurements are as a rule sample dependent, geometry dependent, and excitation source dependent. Thus we compare the *linear* threshold temperature dependence for CdS, CdSe, and ZnO reported by Hvam[33] for 40-kV electron-beam pumping with the *exponential* dependence reported for optical pumping in Ref. 29. The dependence of laser wavelength on temperature also is affected by optical cavity parameters independently of this gain process. Detailed comparison of our results with experimental threshold temperature dependence does not appear useful at this time.

The magnetic field shift observed in $CdSnP_2$ and GaAs is incompatible with free-carrier recombination unless plasma correlation is included, and is also incompatible with the exciton-exciton mechanism. Exciton-phonon, donor-acceptor-pair, exciton-electron, donor-electron–exciton, and bound-exciton processes may also all be excluded when the linewidth, position, and shift with excitation or temperature are considered. The direct recombination including long-range Coulomb interaction is compatible with all the data available, the agreement being in fact quantitative to a degree probably beyond the clear validity of the theory.

Our suggestion that the same mechanism is operative (at least in the case of optical excitation) for stimulated emission in the II-VI compounds and perhaps CuCl as well is not as yet based on direct evidence excluding the exciton-exciton mechanism. The estimated values of r_s in Table I suggest that if a many-body treatment is appropriate for GaAs or $CdSnP_2$, it should be valid to the same degree for all the compounds listed therein. There is indirect evidence consisting of the similarity in behavior of all the direct-gap semiconductors under strong optical excitation which argues for a common explanation of the gain mechanism. Keune and co-workers[10] have pointed out the difficulties associated with exciton-based mechanisms for CdSe; and CdS, ZnO, and the $CdSe_xS_{1-x}$ mixtures exhibit similar if not identical behavior of the stimulated emission characteristics[29,33] when pumped in the same way. (Resolution of the differences between electron-beam pumped and optically pumped spectra awaits more quantitative characterization of the excitation density and distribution resulting from electron-beam pumping. It is possible that electron-beam excitation does not produce excited carrier densities corresponding to $r_s < 5$, and that an interacting exciton description may be appropriate, for instance.) Also, the mechanism discussed here is fully compatible with the observed characteristics of stimulated emission in all the materials in Table I, and can provide the large gains (10^3–10^4 cm^{-1}) measured[4,34] or inferred[29] with optical excitation. Mechanisms involving scattered excitons necessarily have smaller (often much smaller) potential gain, since the direct recombination matrix element is reduced by scattering probability and kinetic factors. We conclude that the direct recombination including exchange and correlation provides the best description of optical gain in optically excited direct-gap semiconductors generally, and is consistent with all the features of the optical gain in such materials known presently.

VI. SUMMARY AND DISCUSSION OF GAIN MEASUREMENTS

We have presented the basis for calculation of optical gain in semiconductors from the point of view of many-body theory. A considerable practical simplification results from replacing the particular with the average self-energy in the Dyson equation for the single-particle electron and hole Green's functions. Nevertheless, numerical calculation is necessary, unless the very simplest form for the inverse dielectric function near the plasma pole is taken, and even then for finite temperature or finite photon wave vector, numerical methods must still be employed. We have emphasized approximately valid analytic expressions and the physical meaning of the results, rather than presenting detailed numerical results here. At least in part we feel that is appropriate because difficulties exist in comparing detailed results to experiment, specifically since the excitation density may not be a well-defined experimental quantity, but also because of difficulties in interpreting experimental gain measurements.

In principle, the small-signal optical gain in semiconductors can be measured as in other lasers.[34] In practice, it is difficult to be confident of the significance of the measurement. It is not now possible to attain uniform optical pumping of direct-gap semiconductors on a quasi-steady-state basis

TABLE I. Estimated values of r_s at threshold for stimulated emission under optical excitation ($T = 77$ °K).

Material	a_0 (10^{-8} cm)	n (cm^{-3})	r_s	Ref.
$In_xGa_{1-x}P$	~120	3×10^{16}	1.7	a
$CdSnP_2$	120	2×10^{17}	0.9	b
GaAs	100	3×10^{16}	2	c
CdSe	54	2×10^{17}	2	d, e
CdS	27	10^{18}	2.3	e
ZnO	14	10^{19}	2.1	e
CuCl	7	10^{20}	2	f

[a] From data in Ref. 6.
[b] From Ref. 7 and J. L. Shay (unpublished).
[c] From D. L. Keune *et al.*, J. Appl. Phys. 42, 2048 (1971).
[d] From Keune *et al.*, Ref. 10.
[e] From data in Ref. 26.
[f] From data in Ref. 4.

throughout a region large enough that diffraction effects[29] are negligible. Measurement of amplified spontaneous emission need not reflect the "true" gain if the spontaneous emission contains features, particularly narrow-line features, unrelated to the principal wideband gain mechanism. The problem is aggravated if the gain saturates for the most part homogeneously. With optical or electron-beam excitation there are invariably regions which are only weakly excited, and in which (at least at low temperatures) narrow-line exciton-process features dominate the spontaneous emission. Amplification of this emission by a broadband-gain process may give data which would appear to indicate narrow-line gain only. In a gas laser,[35] or a dye laser,[36] there are no complications from narrow-line emission processes within the spectral envelope of the gain mechanism of interest and the spontaneous emission may be legitimately used as a "white" test signal. The situation in semiconductor lasers is evidently much more complex. We take the point of view that the gain arises practically from an intrinsic wideband process which appears to be common to all or at least most direct-gap semiconductors, and that appearance of narrow-line gain at the wavelength of some process known in the spontaneous or low-level luminescence is a result similar to the "locking" of a wideband oscillator to a signal generator.

Experiments in which the gain seen by a tunable externally injected signal beam could be measured would be most useful in testing the above suggestion, but these seem almost as difficult as a fully second-quantized theoretical treatment including effects of saturation and spatial variation of excitation density. At present it appears that considerable caution in interpreting experimental results is desirable, particularly with regard to conclusions about the viability of excitons in essentially unchanged form at high excitation density which might appear to follow from the presence of sharp-line features at characteristic wavelengths in the stimulated emission spectra.

Recently, stimulated emission in indirect-gap materials with deep isoelectronic trap complexes has been reported.[37–39] A many-body treatment of the sort presented here is clearly *inappropriate* to apply to this case since the required large excitation density for important screening effects or "Mott transition" of the deep traps is evidently not reached. We expect a predominantly *inhomogeneously* broadened *extrinsic* (i.e., wavelength dependent on chemical nature of isoelectronic trap impurity) gain process to result, as the initial observations indicate is in fact the case.[39]

In conclusion, we have shown that plasma or long-range correlation effects are expected to be important in determining the features of optical gain in semiconductors at the excitation density apparently characteristic of the stimulated regime. In particular, a "blue" shift proportional to the frequency of the dominant plasmon or hybrid plasma mode is expected, in addition to the "red" exchange shift. This prediction is in accord with wavelength shifts arising from modification of the of the plasma frequency via change in external magnetic field. Although theory and experiment are in excellent numerical agreement, there are uncertainties concerning the applicability of the theory which limit the significance of quantitative details. On the other hand, one may use the experimental data to argue that the theory based on random-phase approximation describes at least the gross aspects of the electron-hole plasma important for optical gain in the intermediate excitation-density regime.

ACKNOWLEDGMENTS

It is a pleasure to thank J. L. Shay, A. G. Fox, and W. F. Brinkman of Bell Laboratories, and Professor P. A. Wolff of the Massachusetts Institute of Technology, for helpful discussions and critical comments during the course of this work.

---

[1]Exciton-LO phonon emission has been commonly suggested by many authors for II-VI compounds. The theory for this process was presented in detail by H. Haug, J. Appl. Phys. 39, 4687 (1968).

[2]C. Benoît à la Guillaume, J. Debever, and F. Salvan, Phys. Rev. 177, 567 (1969).

[3]D. Magde and H. Mahr, Phys. Rev. B 2, 4098 (1970).

[4]K. L. Shaklee, R. F. Leheny, and R. E. Nahory, Phys. Rev. Letters 26, 888 (1971).

[5]N. G. Basov, O. V. Bogdankevich, V. A. Goncharov, B. M. Lavrushin, and V. Yu. Sudzilovskii, Dokl. Akad. Nauk SSSR 168, 1283 (1966) [Sov. Phys. Dokl. 11, 522 (1966)].

[6]Cf. R. D. Burnham, N. Holonyak, Jr., D. L. Keune, and D. R. Scifres, Appl. Phys. Letters 18, 160 (1971), and references therein.

[7]J. L. Shay, W. D. Johnston, Jr., E. Buehler, and J. H. Wernick, Phys. Rev. Letters 27, 711 (1971).

[8]J. L. Shay, L. M. Schiavone, E. Buehler, and J. H. Wernick, J. Appl. Phys. 43, 2805 (1972).

[9]J. L. Shay and W. D. Johnston, Jr., this issue, Phys. Rev. B 6, 1605 (1972).

[10]D. L. Keune, N. Holonyak, Jr., P. D. Dapkus, and R. D. Burnham, Appl. Phys. Letters 17, 42 (1970); see also Ref. 6.

[11]J. Billé, Phys. Status Solidi 36, 775 (1969).

[12]H. Aritome, M. Kawabe, K. Masuda, and S. Namba, Sci. Papers Inst. Phys. Chem. Res. (Tokyo) 63, 66 (1969).

[13]J. G. Gay, Phys. Rev. B 4, 2567 (1971). An Erratum is to appear, but the results are qualitatively similar insofar as the purpose of the present paper is concerned. J. G. Gay (private communication).

[14]N. F. Mott, Can. J. Phys. 34, 1356 (1956).
[15]W. F. Brinkman, T. M. Rice, P. W. Anderson, and S. T. Chui, Phys. Rev. Letters 28, 961 (1972).
[16]V. A. Gergel, Fiz. Tverd. Tela 11, 3538 (1969) [Sov. Phys. Solid State 11, 2962 (1970)].
[17]L. V. Keldysh and Yu. V. Kopaev, Fiz. Tverd. Tela 6, 2791 (1964) [Sov. Phys. Solid State 6, 2219 (1965)].
[18]E. O. Kane, J. Phys. Chem. Solids 1, 249 (1957).
[19]V. A. Gergel, R. F. Kazarinov, and R. A. Suris, Zh. Eksperim. i Teor. Fiz. 53, 544 (1967) [Sov. Phys. JETP 26, 354 (1968)].
[20]See, for instance, A. A. Abrikosov, L. P. Gor'Kov, and I. Ye. Dzyaloshinskii, *Quantum Field Theoretical Methods in Statistical Physics*, 2nd ed. (Pergamon, New York, 1965).
[21]We wish only to sketch the standard treatment. The reader interested in details, cautions, etc., is referred to a general text such as A. L. Fetter and J. D. Walecka, *Quantum Theory of Many Particle Systems* (McGraw-Hill, New York, 1971). Our notation should be understood as symbolic only; thus Matsubara sums have been indicated as energy integrals, etc., to avoid undue repetition of well-known details.
[22]B. I. Lundqvist, Physik Kondensierte Materie 6, 193 (1967); 6, 206 (1967); 7, 117 (1968).
[23]N. J. Horing has discussed the utility of the dielectric constant representation as unifying collective and microscopic descriptions of the quantum plasma in *Electronic Structures in Solids*, edited by E. D. Haidemenakis (Plenum, New York, 1969).
[24]This point may appear obvious, and it has received only cursory mention in Ref. 22; nevertheless, we stress it here since it is evidently not universally appreciated [cf. *Note added* to B. D. McCombe, R. J. Wagner, S. Teitler, and I. J. Quinn, Phys. Rev. Letters 28, 38 (1972)].
[25]N. J. Horing, Ann. Phys. (N.Y.) 31, 1 (1965).
[26]N. D. Mermin and E. Canel, Ann. Phys. (N.Y.) 26, 247 (1964).
[27]P. Nozières and D. Pines, Phys. Rev. 111, 442 (1958).
[28]Ya. Pokrovsky, Z. Kaminsky, and K. Svistunova, *Proceedings of the Tenth International Conference on the Physics of Semiconductors* (USAEC Div. Tech. Inf., Springfield, Va., 1970) (Conf-700801, Paper).
[29]W. D. Johnston, Jr., J. Appl. Phys. 42, 2731 (1971).
[30]R. J. Phelan, A. R. Calawa, R. H. Rediker, R. J. Keyes, and B. Lax, Appl. Phys. Letters 3, 143 (1963).
[31]F. L. Galeener, I. Melngailis, G. B. Wright, and R. H. Rediker, J. Appl. Phys. 36, 1574 (1965).
[32]F. L. Galeener, G. B. Wright, W. E. Krag, T. M. Quist, and H. J. Zeiger, Phys. Rev. Letters 10, 472 (1963).
[33]J. M. Hvam, Phys. Rev. B 4, 4459 (1971).
[34]K. L. Shaklee and R. F. Leheny, Appl. Phys. Letters 18, 475 (1971).
[35]W. T. Silfvast and J. S. Deech, Appl. Phys. Letters 11, 97 (1967).
[36]C. V. Shank, A. Dienes, and W. T. Silfvast, Appl. Phys. Letters 17, 307 (1970).
[37]N. Holonyak, Jr., D. R. Scifres, M. G. Craford, W. O. Groves, and D. L. Keune, Appl. Phys. Letters 19, 256 (1971).
[38]R. E. Nahory, K. L. Shaklee, R. F. Leheny, and R. A. Logan, Phys. Rev. Letters 27, 1647 (1971).
[39]N. Holonyak, Jr., D. R. Scifres, H. M. Macksey, R. D. Dupuis, Y. S. Moroz, C. B. Duke, G. G. Kleiman, and F. V. Williams, Phys. Rev. Letters 28, 230 (1972).

PHYSICAL REVIEW B VOLUME 6, NUMBER 4 15 AUGUST 1972

Electronic Band Structure and Optical Properties of 3*C*-SiC, BP, and BN

L. A. Hemstreet, Jr.* and C. Y. Fong
Department of Physics, University of California, Davis, California 95616
(Received 22 February 1972)

The energy band structure and optical properties of the zinc-blende semiconductors 3*C*-SiC, BP, and BN have been calculated using a nonlocal version of the empirical-pseudopotential method. The results of this investigation are discussed and compared to experiment. The agreement between theory and experiment is found to be very good for both SiC and BP. The BN results are quite rough, owing to some very questionable assumptions made necessary because of the scarcity of experimental data. However, the results seem to give a reasonable first approximation to the correct band structure. The effect of the nonlocal p pseudopotential on these three crystals is discussed.

I. INTRODUCTION

During the last several years, the empirical-pseudopotential method (EPM) has been successfully used by many authors to gain valuable insight into the band structure and optical properties of a large group of semiconductors.[1] This method of calculation has been applied to the group-IV elements C, Si, Ge, and α-Sn, which crystallize in the diamond structure, as well as to many of the III-V and II-VI compounds, most of which exhibit the zinc-blende crystal structure. We report here the results of applying the EPM to 3*C*-SiC, BP, and BN. Since the materials discussed in this work are of interest as potential solid-state devices due to their high melting point, chemical inertness,