PHYSICAL REVIEW B VOLUME 24, NUMBER 12 15 DECEMBER 1981

Optical dielectric function for highly excited semiconductors in the moment-conserving approximation

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(Received 23 April 1981)

We have calculated the complex optical dielectric function of an electron-hole plasma in highly excited direct-gap semiconductors using the double-time Green's-function method. The equation of motion for an appropriate Green's function is linearized in the moment-conserving and Hartree-Fock approximations. Their interrelationship and the connection with earlier treatments of the electron-hole correlation is discussed. The electron-hole correlation yields an enhancement of the free-particle response and reproduces the overall shape of the observed gain and absorption spectra satisfactorily. The calculated changes in the refraction index with varying excitation energy exhibit a strong dispersive nonlinearity which explains the occurrence of optical bistability.

I. INTRODUCTION

Recently, there has been considerable interest both experimental[1-3] and theoretical[4-8] in studying the optical response of highly excited direct-gap semiconductors. One creates a system of an electron-hole plasma of variable temperature and density inside a semiconductor crystal by exciting it with a high-power laser beam. It is the optical behavior of this plasma which is under investigation. Transmission experiments give the absorption (gain) line shape from which information about many properties of the system like the electron and hole Fermi energies, their density, and temperature can be inferred. Recent experiments[9] have also shown the existence of optical bistability in such systems. It results from changes in the refractive index which are intensity (and hence density) dependent. Theoretically, the quantity of interest here is the optical dielectric function. Its real and imaginary parts, respectively, give the refractive index and the absorption (gain) spectrum.

It is known that the line shape cannot be accounted for within a noninteracting one-particle model. The experiments show an appreciable enhancement over the free-particle results both in the gain and the absorption region and a considerable influence of collision broadening especially on the long-wavelength side of the gain region. This effect can partly be simulated by the assumption that the momentum is not conserved in the optical transitions. This simple assumption, for which there is, however, no first-principle justification improves the fit in the gain region. At the same time, however, the fit deteriorates[6] for larger energies, i.e., in the absorption region. A better fit is obtained both in the gain and absorption regions if many-body effects such as single-particle energy renormalization, collision broadening, and electron-hole correlation are taken into account.[7] A common feature of the many-body theories[4-8] is to solve the Bethe-Salpeter equation for an appropriate polarization function in varying degrees of sophistication. In all these calculations, only the imaginary part of the dielectric function has been determined. Only recently attempts have been made[10] to calculate the intensity-dependent changes of the refraction index from the corresponding changes in the absorption coefficient close to the band gap.

In this paper,[11] we calculate the full complex optical dielectric function of the system using the double-time Green's function method.[12] We concentrate in the present paper on the treatment of the electron-hole correlation, while we disregard the influence of correlation energy and collision broadening which have already been treated extensively in Ref. 7. In Sec. II, we begin by formulating the problem in terms of the transverse current-density response function which is directly related to the required dielectric function. The equation of motion of an appropriate Green's function is linearized in moment-conserving and Hartree-Fock approximations. The frequency moments are the exact coefficients in the short-time expansion of the response function. Therefore, this approximation shall amount to incorporating the correct high-frequency behavior. The integral equation obtained after decoupling the equation of motion in the Hartree-Fock approximation is solved in two different ways. In the first method, the interaction potential is factorized and we get an expression for the dielectric function which is almost the same as those derived earlier by Zimmerman[5] and Arya and Hanke.[6] The second way is to solve this integral equation variationally. This leads to the same result as obtained in the moment-conserving approximation. The results of our numerical calculation are presented and discussed in Sec. III for the direct-gap semiconductor GaAs.

II. THEORETICAL FORMULATION

An optical probe directly couples to transverse current excitations in a system. Therefore, the response of the system to a space- and time-dependent external light field can be described through a transverse current-density response function. It can be defined[13] in terms of retarded Green's function as

$$\chi(\vec{q},t)=\langle\langle J_t(\vec{q},t);J_t^{\dagger}(\vec{q},0)\rangle\rangle$$
$$=-i\Theta(t)\langle[J_t(\vec{q},t),J_t^{\dagger}(\vec{q},0)]\rangle, \qquad (1)$$

where $\Theta(t)$ is a Heaviside unit step function and the square bracket stands for a commutator. The angular brackets denote a thermal average taken with respect to a grand canonical ensemble defined by the electron-hole plasma Hamiltonian $H=H_0+H'$, H_0 being the single-particle part and H' contains all the interparticle interaction terms in dominant-term approximation. The Fourier transform of the transverse particle-current-density operator is given by

$$J_t(\vec{q},t)=\frac{1}{m}\sum_i(\hat{e}\cdot\vec{p}_i)e^{-i\vec{q}\cdot\vec{r}_i(t)}. \qquad (2)$$

Here, $\hat{e}$ is a unit vector perpendicular to $\vec{q}$; $\vec{r}_i$ and $\vec{p}_i$ are the position and momentum coordinates of an ith particle, respectively. Using Bloch waves as the basis, we write Eq. (2) in the second quantized form as

$$J_t(\vec{q},t)=\sum_{\vec{k},n,n'}M_{nn'}(\vec{k},\vec{k}+\vec{q})$$
$$\times a^{\dagger}_{\vec{k},n}(t)a_{\vec{k}+\vec{q},n'}(t), \qquad (3)$$

where n and n' run over different valence and conduction bands. $M_{nn'}(\vec{k},\vec{k}+\vec{q})$ is the transition matrix element

$$M_{nn'}(\vec{k},\vec{k}+\vec{q})=\hat{e}\cdot\int_{\text{cell}}d^3r\,U^{*}_{\vec{k},n}(\vec{r})$$
$$\times\left(-\frac{i\vec{\nabla}}{m}\right)U_{\vec{k}+\vec{q},n'}(\vec{r}), \qquad (4)$$

$U_{\vec{k},n}(\vec{r})$ being the cell periodic part of the usual Bloch wave function. Further, $a^{\dagger}_{\vec{k}n}(a_{\vec{k}n})$ is a particle creation (annihilation) operator in band n. In Eq. (3) and thereafter, particle spin is considered to be included in its wave-vector index and $\hbar$ is taken to be equal to unity.

In order to simplify expression (3), we make use of the familiar two-band approximation, i.e., all the valence electrons in a nonexcited crystal are assumed to be in the topmost valence band which on excitation can jump to the lowest conduction band. Also the photon wave vector $\vec{q}$ can be safely neglected in comparison to the linear dimensions of a Brillouin zone. Making use of these approximations in Eq. (3) and confining only to the interband current, we obtain

$$J_t(\vec{q},t)=J(t)=\sum_{\vec{k}}[M_{vc}(\vec{k})b_{-\vec{k}}(t)a_{\vec{k}}(t)$$
$$+M^{*}_{vc}(\vec{k})a^{\dagger}_{\vec{k}}(t)b^{\dagger}_{-\vec{k}}(t)], \qquad (5)$$

where $a_{\vec{k}}$ and $b_{\vec{k}}$ now refer to electron and hole operators, respectively.

In view of Eq. (5), expression (1) for the current-density response function becomes

$$\chi(t)\equiv\sum_{\vec{k}_1,\vec{k}_2}\langle\langle[M_{vc}(\vec{k}_1)b_{-\vec{k}_1}(t)a_{\vec{k}_1}(t)+M^{*}_{vc}(\vec{k}_1)a^{\dagger}_{\vec{k}_1}(t)b^{\dagger}_{-\vec{k}_1}(t)];[M^{*}_{vc}(\vec{k}_2)a^{\dagger}_{\vec{k}_2}(0)b^{\dagger}_{-\vec{k}_2}(0)+M_{vc}(\vec{k}_2)b_{-\vec{k}_2}(0)a_{\vec{k}_2}(0)]\rangle\rangle\,. \qquad (6)$$

If an equation of motion is written for the full Green's function as such, then it is not possible even to separate the free-particle part (i.e., corresponding to H_0 part of the Hamiltonian). We rewrite Eq. (6) in two parts as

$$\chi(t)=\sum_{\vec{k}_1,\vec{k}_2}[M_{vc}(\vec{k}_1)\,\langle\langle b_{-\vec{k}_1}(t)a_{\vec{k}_1}(t);M^{*}_{vc}(\vec{k}_2)a^{\dagger}_{\vec{k}_2}b^{\dagger}_{-\vec{k}_2}+M_{vc}(\vec{k}_2)b_{-\vec{k}_2}a_{\vec{k}_2}\rangle\rangle$$
$$+M^{*}_{vc}(\vec{k}_1)\,\langle\langle a^{\dagger}_{\vec{k}_1}(t)b^{\dagger}_{-\vec{k}_1}(t);M^{*}_{vc}(\vec{k}_2)a^{\dagger}_{\vec{k}_2}b^{\dagger}_{-\vec{k}_2}+M_{vc}(\vec{k}_2)b_{-\vec{k}_2}a_{\vec{k}_2}\rangle\rangle]\,. \qquad (7)$$

As the correlation of electron or hole operators in the same space (i.e., either creation or annihilation) is zero, Eq. (7) simplifies to

$$\chi(t)=\chi_1(t)+\chi_2(t), \qquad (8a)$$

where

$$\chi_1(t)=\sum_{\vec{k}_1,\vec{k}_2}M_{vc}(\vec{k}_1)M^{*}_{vc}(\vec{k}_2)\,\langle\langle b_{-\vec{k}_1}(t)a_{\vec{k}_1}(t);a^{\dagger}_{\vec{k}_2}b^{\dagger}_{-\vec{k}_2}\rangle\rangle \qquad (8b)$$

$$\chi_2(t)=\sum_{\vec{k}_1,\vec{k}_2} M^*_{vc}(\vec{k}_1)M_{vc}(\vec{k}_2)\langle\langle a^\dagger_{\vec{k}_1}(t)b^\dagger_{-\vec{k}_1}(t);b^\dagger_{-\vec{k}_2}a_{\vec{k}_2}\rangle\rangle. \tag{8c}$$

The optical dielectric function, which contains all the information about the optical properties of the system is given by

$$\epsilon(\omega)=\epsilon_\infty-\frac{4\pi e^2}{\omega^2}\chi(\omega), \tag{9}$$

where ϵ_∞ is the high-frequency optical dielectric constant of the crystal.

In order to calculate $\chi(\omega)$, we introduce appropriate Green's functions through

$$\chi_1(t)=\sum_{\vec{k}_1,\vec{k}_2} M_{vc}(\vec{k}_1)M^*_{vc}(\vec{k}_2)G_1(\vec{k}_1,\vec{k}_2,t), \tag{10a}$$

$$G_1(\vec{k}_1,\vec{k}_2,t)=\langle\langle b_{-\vec{k}_1}(t)a_{\vec{k}_1}(t);\ a^\dagger_{\vec{k}_2}b^\dagger_{-\vec{k}_2}\rangle\rangle, \tag{10b}$$

and

$$\chi_2(t)=\sum_{\vec{k}_1,\vec{k}_2} M^*_{vc}(\vec{k}_1)M_{vc}(\vec{k}_2)G_2(\vec{k}_1,\vec{k}_2,t), \tag{10c}$$

$$G_2(\vec{k}_1,\vec{k}_2,t)=\langle\langle a^\dagger_{\vec{k}_1}(t)b^\dagger_{-\vec{k}_1}(t);\ b^\dagger_{-\vec{k}_2}a_{\vec{k}_2}\rangle\rangle. \tag{10d}$$

The equation of motion for the Green's function $G_1(\vec{k}_1,\vec{k}_2,t)$ is given by

$$i\frac{d}{dt}G_1(\vec{k}_1,\vec{k}_2,t)=-\delta(t)\delta_{\vec{k}_1,\vec{k}_2}[n_e(\vec{k}_1)+n_h(\vec{k}_1)-1] +\langle\langle[b_{-\vec{k}_1}(t\,a_{\vec{k}_1}(t),H'];\ a^\dagger_{\vec{k}_2}b^\dagger_{-\vec{k}_2}\rangle\rangle, \tag{11}$$

where $n_e(\vec{k}_1)$ and $n_h(\vec{k}_1)$ are the electron and the hole Fermi distribution functions, respectively. The Fourier transform of the Green's function as obtained from Eq. (11) is given by

$$G_1(\vec{k}_1,\vec{k}_2,\omega)=-\frac{n_e(\vec{k}_1)+n_h(\vec{k}_1)-1}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}\delta_{\vec{k}_1,\vec{k}_2} +\frac{\langle\langle[b_{-\vec{k}_1}(t)a_{\vec{k}_1}(t),H'];\ a^\dagger_{\vec{k}_2}b^\dagger_{-\vec{k}_2}\rangle\rangle_\omega}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta} \tag{12}$$

where $\epsilon_e(\vec{k}_1)[\epsilon_h(\vec{k}_1)]$ is the band energy of an electron (hole) and η is a positive infinitesimal quantity. In a similar manner, we write the expression for the Fourier transform of Green's function $G_2(\vec{k}_1,\vec{k}_2,t)$ as

$$G_2(\vec{k}_1,\vec{k}_2,\omega)=\frac{n_e(\vec{k}_1)+n_h(\vec{k}_1)-1}{\omega+[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}\delta_{\vec{k}_1,\vec{k}_2} +\frac{\langle\langle[a^\dagger_{\vec{k}_1}(t)b^\dagger_{-\vec{k}_1}(t),H'];b_{-\vec{k}_2}a_{\vec{k}_2}\rangle\rangle_\omega}{\omega+[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}. \tag{13}$$

It is seen from Eqs. (12) and (13) that the nonresonant term $G_2(\vec{k}_1,\vec{k}_2,\omega)$ is much smaller than the resonant term $G_1(\vec{k}_1,\vec{k}_2,\omega)$. Therefore, we neglect $\chi_2(\omega)$ in comparison to $\chi_1(\omega)$ in the calculation of the full response function $\chi(\omega)$.

The Eq. (12) for the two-particle Green's function contains a three-particle Green's function. We shall decouple Eq. (12) using approximations described below.

A. Moment conserving decoupling approximation

In this section we shall linearize Eq. (12) making moment-conserving approximations. It amounts to incorporating the high-frequency behavior of the response function correctly as frequency moments are its expansion coefficients in the corresponding limit. Using Eqs. (8), (10), and (12), we first write the expression for the interband current response function in the form

$$\chi(\omega)=\chi_0(\omega)+\sum_{\vec{k}_1}\frac{1}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}\sum_{\vec{k}_2}M_{vc}(\vec{k}_1)M^*_{vc}(\vec{k}_2)\langle\langle[b_{-\vec{k}_1}(t)a_{\vec{k}_1}(t),H'];a^\dagger_{\vec{k}_2}b^\dagger_{-\vec{k}_2}\rangle\rangle_\omega. \tag{14}$$

Here

$$\chi_0(\omega)=-\sum_{\vec{k}_1}|M_{vc}(\vec{k}_1)|^2\frac{n_e(\vec{k}_1)+n_h(\vec{k}_1)-1}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}, \tag{15}$$

gives the response of the noninteracting electrons and holes.

1. Conserving first moment

We write the higher-order Green's function in Eq. (14) in terms of lower order one by making use of an ansatz[14]

$$\sum_{\vec{k}_2}M_{vc}(\vec{k}_1)M^*_{vc}(\vec{k}_2)\langle\langle[b_{-\vec{k}_1}(t)a_{\vec{k}_1}(t),H'];\ a^\dagger_{\vec{k}_2}b^\dagger_{-\vec{k}_2}\rangle\rangle_\omega=A_{\vec{k}_1}\sum_{\vec{k}_3,\vec{k}_2}M_{vc}(\vec{k}_3)M^*_{vc}(\vec{k}_2)\langle\langle b_{-\vec{k}_3}(t)a_{\vec{k}_3}(t);a^\dagger_{\vec{k}_2}b^\dagger_{-\vec{k}_2}\rangle\rangle_\omega, \tag{16}$$

where for simplicity, the coefficient $A_{\vec{k}_1}$ is considered to be frequency independent. Equations (14) and (16) together give

$$\chi(\omega)=\frac{\chi_0(\omega)}{1-F(\omega)}\,, \tag{17}$$

where

$$F(\omega)=\sum_{\vec{k}_1}\frac{A_{\vec{k}_1}}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta} \tag{18}$$

takes into account the effect of electron-hole correlations in the system. The coefficient $A_{\vec{k}_1}$ is determined by equating the first frequency moment of both sides in Eq. (16). It leads to

$$A_{\vec{k}_1}=\frac{1}{\langle\omega^1\rangle}\sum_{\vec{k}_2}M_{vc}(\vec{k}_1)M^*_{vc}(\vec{k}_2)\langle[[[b_{-\vec{k}_1}a_{\vec{k}_1},H'],H],a^\dagger_{\vec{k}_2}b^\dagger_{-\vec{k}_2}]\rangle\,, \tag{19a}$$

where

$$\langle\omega^1\rangle=\sum_{\vec{k}_1,\vec{k}_2}M_{vc}(\vec{k}_1)M^*_{vc}(\vec{k}_2)\langle[[b_{-\vec{k}_1}a_{\vec{k}_1},H],a^\dagger_{\vec{k}_2}b^\dagger_{-\vec{k}_2}]\rangle\,, \tag{19b}$$

is the first frequency moment of $\chi_1(\omega)$. The commutators involved in Eqs. (19a and (19b) are solved in a straightforward manner. The expressions still involve averages of the product of four (fermion) operators which are decoupled using the familiar Hartree-Fock approximation, which for a typical term may be written as

$$\langle a^\dagger_{\vec{k}_1}b^\dagger_{-\vec{k}_2}a_{\vec{k}_3}b_{-\vec{k}_4}\rangle=-\langle a^\dagger_{\vec{k}_1}a_{\vec{k}_3}\rangle\langle b^\dagger_{-\vec{k}_2}b_{-\vec{k}_4}\rangle\,. \tag{20}$$

Finally, we get the following expression for the local field term:

$$\begin{aligned}F(\omega)=-\frac{1}{\langle\omega^1\rangle}\sum_{\vec{k}_1,\vec{k}_2}V(\vec{k}_2-\vec{k}_1)M_{vc}(\vec{k}_1)&\frac{n_e(\vec{k}_1)+n_h(\vec{k}_1)-1}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}\\ &\times\{M^*_{vc}(\vec{k}_2)[\epsilon_e(\vec{k}_2)+\epsilon_h(\vec{k}_2)][n_e(\vec{k}_2)+n_h(\vec{k}_2)-1]\\ &\quad-M^*_{vc}(\vec{k}_1)[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)][n_e(\vec{k}_2)+n_h(\vec{k}_2)]\}\,,\end{aligned} \tag{21}$$

and the first moment

$$\begin{aligned}\langle\omega^1\rangle=&-\sum_{\vec{k}_1}|M_{vc}(\vec{k}_1)|^2[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)][n_e(\vec{k}_1)+n_h(\vec{k}_1)-1]\\ &-\sum_{\vec{k}_1,\vec{k}_2}V(\vec{k}_2-\vec{k}_1)M_{vc}(\vec{k}_1)[n_e(\vec{k}_1)+n_h(\vec{k}_1)-1]\\ &\qquad\times\{M^*_{vc}(\vec{k}_2)[n_e(\vec{k}_2)+n_h(\vec{k}_2)-1]-M^*_{vc}(\vec{k}_1)[n_e(\vec{k}_2)+n_h(\vec{k}_2)]\}\,.\end{aligned} \tag{22}$$

2. *Generalization of approximation (16)*

It is possible to generalize the decoupling approximation (16) by making the proportionality coefficient frequency dependent,[15] i.e.,

$$\begin{aligned}\sum_{\vec{k}_2}M_{vc}(\vec{k}_1)M^*_{vc}(\vec{k}_2)\langle\langle[b_{-\vec{k}_1}(t)a_{\vec{k}_1}(t),H'];a^\dagger_{\vec{k}_2}b^\dagger_{-\vec{k}_2}\rangle\rangle_\omega&\\ =A_{\vec{k}_1}(\omega)\chi_1(\omega)\,.&\end{aligned} \tag{23}$$

To calculate $A_{\vec{k}_1}(\omega)$, we write the above equation as

$$A_{\vec{k}_1}(\omega)=\frac{\int_{-\infty}^{\infty}dt\,K(\vec{k}_1,t)e^{i(\omega+i\eta)t}}{\int_{-\infty}^{\infty}dt\,\chi_1(t)e^{i\ \omega+i\eta)t}} \tag{24}$$

where

$$K(\vec{k}_1,t)=-i\Theta(t)\sum_{\vec{k}_2}M_{vc}(\vec{k}_1)M^*_{vc}(\vec{k}_2)\langle[[b_{-\vec{k}_1}(t)a_{\vec{k}_1}(t),H'],a^\dagger_{\vec{k}_2}b^\dagger_{-\vec{k}_2}]\rangle\,. \tag{25}$$

Evaluation of $A_{\vec{k}_1}(\omega)$ involves the calculation of averages of the commutators of the type $[B(t), C(0)]$. To calculate them, we first get equal time commutators using the familiar expansion of a time-dependent Heisenberg operator

$$B(t)=e^{iHt}B(0)e^{-iHt}=\sum_{n=0}^{\infty}\frac{(it)^n}{n!}[H,[H,\ldots[H,B(0)]\ldots]]\,. \tag{26}$$

Then in order to get manageable expressions, H is replaced by H_0 in such expansions [however the effect of interactions in $A_{\vec{k}_1}(\omega)$ is there to first order through H' which appears explicitly in its numerator]. Still averages of the products of four operators are involved which are evaluated using Eq. (20). After this is done, the result obtained for $\langle[B(t),C(0)]\rangle$ is in the form of geometric series which can be easily summed up. The result obtained is

$$\begin{aligned}K(\vec{k}_1,t)=i\Theta(t)\sum_{\vec{k}_2}V(\vec{k}_2)M_{vc}(\vec{k}_1)[n_e(\vec{k}_1)+n_h(\vec{k}_1)-1]\\ \times(M^*_{vc}(\vec{k}_1+\vec{k}_2)[n_e(\vec{k}_1+\vec{k}_2)+n_h(\vec{k}_1+\vec{k}_2)-1]\exp\{-i[\epsilon_e(\vec{k}_1+\vec{k}_2)+\epsilon_h(\vec{k}_1+\vec{k}_2)]t\}\\ -M^*_{vc}(\vec{k}_1)[n_e(\vec{k}_1+\vec{k}_2)+n_h(\vec{k}_1+\vec{k}_2)]\exp\{-i[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]t\})\,,\end{aligned} \tag{27}$$

and

$$\chi_1(t)=i\Theta(t)\sum_{\vec{k}_2}|M_{vc}(\vec{k}_2)|^2[n_e(\vec{k}_2)+n_h(\vec{k}_2)-1]\exp\{-i[\epsilon_e(\vec{k}_2)+\epsilon_h(\vec{k}_2)]t\}\,. \tag{28}$$

Using Eqs. (27) and (28) in Eq. (24), we obtain

$$\begin{aligned}A_{\vec{k}_1}(\omega)=-\frac{1}{\chi_0(\omega)}\sum_{\vec{k}_2}V(\vec{k}_2-\vec{k}_1)M_{vc}(\vec{k}_1)[n_e(\vec{k}_1)+n_h(\vec{k}_1)-1]\\ \times\left(M^*_{vc}(\vec{k}_2)\frac{n_e(\vec{k}_2)+n_h(\vec{k}_2)-1}{\omega-[\epsilon_e(\vec{k}_2)+\epsilon_h(\vec{k}_2)]+i\eta}-M^*_{vc}(\vec{k}_1)\frac{n_e(\vec{k}_2)+n_h(\vec{k}_2)}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}\right).\end{aligned} \tag{29}$$

The corresponding expression for the generalized local-field factor as obtained from Eqs. (18) and (29) is given by

$$\begin{aligned}F(\omega)=-\frac{1}{\chi_0(\omega)}\sum_{\vec{k}_1,\vec{k}_2}V(\vec{k}_2-\vec{k}_1)M_{vc}(\vec{k}_1)\frac{n_e(\vec{k}_1)+n_h(\vec{k}_1)-1}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}\\ \times\left(M^*_{vc}(\vec{k}_2)\frac{n_e(\vec{k}_2)+n_h(\vec{k}_2)-1}{\omega-[\epsilon_e(\vec{k}_2)+\epsilon_h(\vec{k}_2)]+i\eta}-M^*_{vc}(\vec{k}_1)\frac{n_e(\vec{k}_2)+n_h(\vec{k}_2)}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}\right).\end{aligned} \tag{30}$$

It is interesting to see if one can reproduce the result (21) obtained by conserving the first moment sum rule from the present approximation. For that, we define a frequency-independent coefficient

$$A_{\vec{k}_1}=\frac{K(\vec{k}_1,t)}{\chi_1(t)}\,, \tag{31}$$

and retain[15] terms proportional to t only in Eqs. (27) and (28). It gives

$$\begin{aligned}A_{\vec{k}_1}=-\frac{1}{\langle\omega^1\rangle_0}\sum_{\vec{k}_2}V(\vec{k}_2-\vec{k}_1)M_{vc}(\vec{k}_1)[n_e(\vec{k}_1)+n_h(\vec{k}_1)-1]\\ \times\{M^*_{vc}(\vec{k}_2)[\epsilon_e(\vec{k}_2)+\epsilon_h(\vec{k}_2)][n_e(\vec{k}_2)+n_h(\vec{k}_2)-1]\\ -M^*_{vc}(\vec{k}_1)[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)][n_e(\vec{k}_2)+n_h(\vec{k}_2)]\}\,,\end{aligned} \tag{32}$$

where $\langle\omega^1\rangle_0$ is the first moment evaluated in the free-particle approximation, i.e., the one given by only the first term in Eq. (22). When Eq. (32) is used in Eq. (18), we get a result for $F(\omega)$ which is not the same as given by Eq. (21). Rather it involves $\langle\omega^1\rangle_0$ while it is the exact first moment which appears in Eq. (21). This is in contrast to what has been found in the study of density[15] and spin density[16] correlations in interacting electron gas. There, two such results coincided as the contribution of the interaction term in the Hamiltonian to the first moment of the density and the spin-density response functions is zero. It is not so in the present case as we are dealing with current correlations.

B. Hartree-Fock approximation

Another simple approximation to linearize Eq. (12) is to use directly Eq. (20) for decoupling the averages of the products of four electron-hole operators in its last term. This leads to an integral equation

$$G_1(\vec{k}_1,\vec{k}_2,\omega) = \tilde{G}_1^0(\vec{k}_1,\omega) \times \Big(\delta_{\vec{k}_1,\vec{k}_2} - \sum_{\vec{k}_3} V(\vec{k}_3-\vec{k}_1)G_1(\vec{k}_3,\vec{k}_2,\omega)\Big), \qquad (33)$$

where

$$\tilde{G}_1^0(\vec{k}_1,\omega) = -\frac{n_e(\vec{k}_1)+n_h(\vec{k}_1)-1}{\omega-[\tilde{\epsilon}_e(\vec{k}_1)+\tilde{\epsilon}_h(\vec{k}_1)]+i\eta}. \qquad (34)$$

Here, $\tilde{\epsilon}_e(\vec{k}_1)$ and $\tilde{\epsilon}_h(\vec{k}_1)$ are the single electron and hole energies, respectively, including the effect of exchange self-energy, i.e.,

$$\tilde{\epsilon}_{e,h}(\vec{k}_1) = \epsilon_{e,h}(\vec{k}_1) - \sum_{\vec{k}_2} V(\vec{k}_2-\vec{k}_1)n_{e,h}(\vec{k}_2). \qquad (35)$$

Now, we shall discuss briefly the solution of Eq. (33) in two different approximations.

1. By factorization of interaction potential

In this subsection, we shall assume that the Coulomb potential in Eq. (33) is statically screened. Then it is very easy to solve this integral equation by factorizing[17] the interaction potential as

$$V(\vec{k}_3-\vec{k}_1) = \frac{V(k_3,k^*)V(k_1,k^*)}{V(k^*,k^*)}. \qquad (36)$$

Here, $V(k_3,k_1)$ is the angular average of $V(\vec{k}_3-\vec{k}_1)$ and k^* is defined by

$$\omega-[\tilde{\epsilon}_e(k^*)+\tilde{\epsilon}_h(k^*)]=0. \qquad (37)$$

Using Eq. (36) in Eq. (33) and further using the Fourier transforms of Eqs. (8a) and (10a), we obtain for the interband current response function

$$\chi(\omega) = \tilde{\chi}_0(\omega) - \frac{\sum_{\vec{k}_1} M_{vc}(k_1)V(k_1,k^*)\tilde{G}_1^0(\vec{k}_1,\omega)\sum_{\vec{k}_2} M_{vc}^*(\vec{k}_2)V(k_2,k^*)\tilde{G}_1^0(\vec{k}_2,\omega)}{V(k^*,k^*)+\sum_{\vec{k}_1} V^2(k_1,k^*)\tilde{G}_1^0(\vec{k}_1,\omega)} \qquad (38)$$

In Eq. (38)

$$\tilde{\chi}_0(\omega) = \sum_{\vec{k}_1} |M_{vc}(\vec{k}_1)|^2\tilde{G}_1^0(\vec{k}_1,\omega) \qquad (39)$$

describes the response of the noninteracting system except that the single-particle energies are renormalized by Hartree-Fock self-energies. The effect of electron-hole correlations are explicitly contained in the second term of Eq. (38). In order to make connection with earlier work, we take the transition matrix elements to be constant for the moment. Then using Eq. (38) in Eq. (9), the expression obtained for the imaginary part of the dielectric function, which directly gives the absorption (gain) spectrum of the system, can be put in the form

$$\epsilon_2(\omega) = -\frac{4\pi e^2}{\omega^2}\tilde{\chi}_0''(\omega)\tilde{\rho}(\omega), \qquad (40)$$

where the enhancement function is

$$\tilde{\rho}(\omega) = \left|\frac{1+\tilde{I}(\omega)-\tilde{J}(\omega)}{1+\tilde{I}(\omega)}\right|^2. \qquad (41)$$

Here

$$\tilde{J}(\omega) = \sum_{\vec{k}} V(k,k^*)\tilde{G}_1^0(\vec{k},\omega) \qquad (42a)$$

and

$$\tilde{I}(\omega) = \frac{1}{V(k^*,k^*)}\sum_{\vec{k}} V^2(k,k^*)\tilde{G}_1^0(\vec{k},\omega). \qquad (42b)$$

It is easily seen that the result (40) is the same as that earlier obtained by Zimmerman[5] and Arya and Hanke[6] except for a difference in self-energies. This difference amounts to having a differently renormalized band gap and does not affect the line shapes.

2. By a variational method

It is possible to solve integral equation (33) using a variational method already used by Rajagopal[18] and Langreth.[19] For that we introduce a vertex function through

$$G_1(\vec{k},\vec{k}_2,\omega)=-\frac{n_e(\vec{k}_1)+n_h(\vec{k}_1)-1}{\omega-[\tilde{\epsilon}_e(\vec{k}_1)+\tilde{\epsilon}_h(\vec{k}_1)]+i\eta}\,\delta_{\vec{k}_1,\vec{k}_2}\Gamma(\vec{k}_1,\omega)\,, \tag{43}$$

so that

$$\chi(\omega)=-\sum_{\vec{k}_1}|M_{vc}(\vec{k}_1)|^2\frac{n_e(\vec{k}_1)+n_h(\vec{k}_1)-1}{\omega-[\tilde{\epsilon}_e(\vec{k}_1)+\tilde{\epsilon}_h(\vec{k}_1)]+i\eta}\,\Gamma(\vec{k}_1,\omega)\,. \tag{44}$$

Also using Eq. (43) in Eq. (33), we obtain

$$\chi(\omega)=\tilde{\chi}_0(\omega)-\sum_{\vec{k}_1,\vec{k}_2}V(\vec{k}_2-\vec{k}_1)M_{vc}(\vec{k}_1)M^*_{vc}(\vec{k}_2)\times\left(\frac{n_e(\vec{k}_1)+n_h(\vec{k}_1)-1}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}\right)\left(\frac{n_e(\vec{k}_2)+n_h(\vec{k}_2)-1}{\omega-[\epsilon_e(\vec{k}_2)+\epsilon_h(\vec{k}_2)]+i\eta}\right)\Gamma(\vec{k}_2,\omega)\,. \tag{45}$$

One can easily construct a Lagrangian of $\Gamma(\vec{k}_1,\omega)$ and find that the simplest variational solution is the one where the vertex function is independent of $\vec{k}$. Then we get from Eqs. (44) and (45)

$$\Gamma(\omega)=\frac{1}{1-F_1(\omega)}\,, \tag{46}$$

and thus

$$\chi(\omega)=\frac{\tilde{\chi}_0(\omega)}{1-F_1(\omega)}\,, \tag{47}$$

where

$$F_1(\omega)=-\frac{1}{\tilde{\chi}_0(\omega)}\sum_{\vec{k}_1,\vec{k}_2}V(\vec{k}_2-\vec{k}_1)M_{vc}(\vec{k}_1)M^*_{vc}(\vec{k}_2)\times\left(\frac{n_e(\vec{k}_1)+n_h(\vec{k}_1)-1}{\omega-[\tilde{\epsilon}_e(\vec{k}_1)+\tilde{\epsilon}_h(\vec{k}_1)]+i\eta}\right)\left(\frac{n_e(\vec{k}_2)+n_h(\vec{k}_2)-1}{\omega-[\tilde{\epsilon}_e(\vec{k}_2)+\tilde{\epsilon}_h(\vec{k}_2)]+i\eta}\right). \qquad (48$$

Insight about the relation between the results which have been obtained by the variational method and by the moment-conserving approximation (30) can be obtained by using a slightly modified formulation of the variational method. For that we rewrite Eq. (33) by writing the self-energy terms explicitly, i.e.,

$$G_1(\vec{k}_1,\vec{k}_2,\omega)=G_1^0(\vec{k}_1,\omega)\left(\delta_{\vec{k}_1,\vec{k}_2}-\sum_{\vec{k}_3}V(\vec{k}_3-\vec{k}_1)G_1(\vec{k}_3,\vec{k}_2,\omega)\right)+\sum_{\vec{k}_3}V(\vec{k}_3-\vec{k}_1)\frac{n_e(\vec{k}_3)+n_h(\vec{k}_3)}{\omega-[\epsilon_e(\vec{k}_1)+\epsilon_h(\vec{k}_1)]+i\eta}\,G_1(\vec{k}_1,\vec{k}_2,\omega)\,. \tag{49}$$

Solving this equation again for a constant vertex function,[20] we recover exactly the result (17), where the local-field correction is given by Eq. (30). Thus, one sees that the second version of the variational method and the moment-conserving approximation yield identical results.[21] Comparing the two variational solutions given by Eqs. (30) and (48), we see that the second term in the large parentheses of Eq. (30) is due to the self-energy corrections of the single-particle energies. Since it is known from experiments, that band-gap renormalizations are very important, one has to take a formulation in which renormalized single-particle energies are used [as it is done in Eq. (48)].

For the following numerical evaluations we shall use the result (21), which has been derived by conserving the first moment. A comparison between these equations and Eq. (30) shows that the second terms in the curly brackets of Eqs. (21) and (22), respectively, are again due to self-energy corrections. These terms have to be left out if renormalized single-particle energies are to be used.

III. NUMERICAL RESULTS AND DISCUSSION

We now evaluate our expressions numerically for GaAs and compare the results with experiments. We need to calculate the real and imaginary parts of the free-particle response and the local-field factor. For that, as can be seen from the corresponding equations, full band-structure information is needed in order to know the transition matrix elements and the electron-hole band energies as functions of wave vector over the whole Brillouin zone. In the present paper we confine ourselves to a simplified model calculation in which the transition matrix elements are taken to be constant—their value is determined by constraining the calculated maximum gain value to be the same as experimentally observed. Further, for band energies, the effective-mass approximation is used near the center of the zone. For large wave vectors, tight-binding expressions corresponding to simple cubic lattice are used. The density of states for such energy bands is finite at all points inside or at the Brillouin zone which is not so, e.g., for analogous expressions for face centered lattice. Jelitto[22] has accurately fitted the actual density of states by a simple algebraic function. We need joint density of states for electrons and holes which follows trivially from his expression.

Keeping in view the above simplifying assumptions for the actual band structure, we confine ourselves to the evaluation of the dielectric function according to our simplest approximation (21) with the modifications mentioned at the end of the preceding section. The material parameters used are $m_e = 0.067\,m_0$, $m_h = 0.54\,m_0$, $\epsilon_\infty = 10.9$. The effective bandwidth of the conduction and valence bands is taken as 2.15 eV. The self-energy corrections are taken within the rigid-band-shift approximation and the frequency is measured with respect to the experimental value of renormalized band gap E_g'. We calculate simultaneously the real $[\epsilon_1(\omega)]$ and the imaginary $[\epsilon_2(\omega)]$ part of the optical dielectric function. It is then straightforward to calculate the refraction index $n'(\omega)$ and the absorption coefficient $\alpha(\omega)$ as

$$n'(\omega) = \left(\tfrac{1}{2}\{\epsilon_1(\omega) + [\epsilon_1^2(\omega) + \epsilon_2^2(\omega)]^{1/2}\}\right)^{1/2},$$

and

$$\alpha(\omega) = \frac{\omega}{n'(\omega)c}\,\epsilon_2(\omega)\,.$$

Figure 1 shows a fit of the resulting absorption spectrum to the experiment,[1] together with the real part $\epsilon_1(\omega)$. The resulting plasma temperature is 10 K and the plasma density is 5.3×10^{16} cm^{-3}. These parameters are quite close to those obtained in a theory of the absorption spectrum[7] which also contained the effects of collision broadening, but did not allow the direct calculation of $\epsilon_1(\omega)$ because the calculation of $\alpha(\omega)$ was limited to the band edge region. The collision broadening gives rise to a tail on the low-frequency side of the gain spectrum. This effect is not taken into account in the present treatment. The excitonic enhancement due to the local-field correction $F(\omega)$ is clearly seen at frequencies around the quasichemical potential μ, which determines the crossing from gain to absorption. The change in the absorption

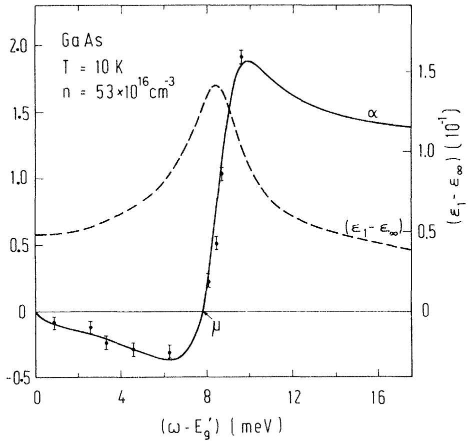

FIG. 1. Absorption spectrum $\alpha(\omega)$ (full line) and the spectrum of the real part of the dielectric function $\epsilon_1(\omega)-\epsilon_\infty$ (dashed line) for GaAs with a plasma density $n = 5.3\times10^{16}$ cm^{-3} and a plasma temperature $T = 10$ K. The experimental points are taken from Ref. 1.

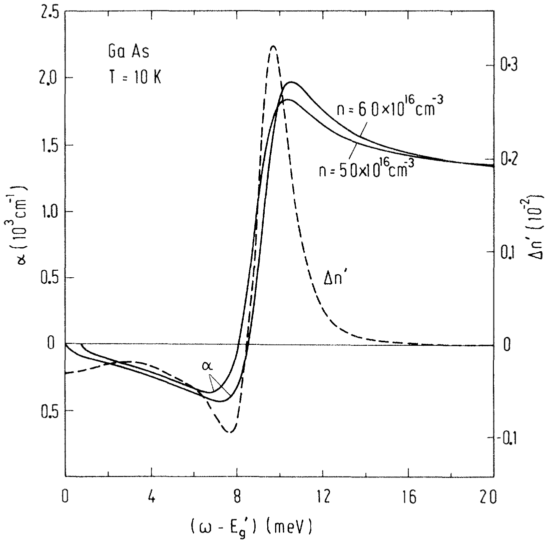

FIG. 2. Two absorption spectra (full lines) for GaAs at plasma densities of 5×10^{16} and 6×10^{16} cm^{-3} at $T = 10$ K. The corresponding change in the refraction index $\Delta n'(\omega)$ is shown by the dashed curve.

spectrum for two close plasma densities which can be obtained, e.g., using slightly different excitation intensities, is shown in Fig. 2, where $\alpha(\omega)$ is plotted for plasma densities of 5×10^{16} and 6×10^{16} cm^{-3}. The density dependence of the renormalized band gap E_g' is taken into account according to Ref. 1. The corresponding change of the refraction index for the small change in plasma density is also plotted. It shows a strong frequency dependence in the neighborhood of the quasichemical potential. This large dispersive nonlinearity causes the occurrence of optical bistability.[9] The resulting spectrum of $\Delta n'(\omega)$ is similar to that obtained[10] by a Kramers-Kronig transformation of the corresponding changes $\Delta\alpha(\omega)$, where the absorption spectra have been calculated[7] in the effective-mass approximation. Detailed measurements of the excitation intensity-dependent spectrum of the refraction index are not yet available.

ACKNOWLEDGMENTS

This work is a project of the Sonderforschungsbereich Frankfurt/Darmstadt financed by special funds of the Deutsche Forschungsgemeinschaft. One of us (H.B.S.) would like to thank Professor M. P. Tosi for useful discussions.

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