Nonequilibrium theory of the optical Stark effect and spectral hole burning in semiconductors

S. Schmitt-Rink
AT&T Bell Laboratories, Murray Hill, New Jersey 07974-2070

D. S. Chemla
AT&T Bell Laboratories, Holmdel, New Jersey 07733

H. Haug
Institut für Theoretische Physik, Universität Frankfurt, Robert-Mayer-Strasse 8, D-6000 Frankfurt am Main, Federal Republic of Germany

(Received 14 July 1987)

We consider the influence of intense coherent laser fields on the electronic and optical properties of semiconductors. Using nonequilibrium Green's-function techniques and exploiting the analogies to superconducting and Bose-condensed systems, we discuss the nature of the renormalizations and the collective excitations in the collisionless regime. Experimentally, this situation can be realized (i) under nonresonant excitation of virtual electron-hole pairs and (ii) under resonant excitation with ultrashort pulses. We explain the recently observed optical Stark effect as well as spectral hole burning and derive from first principles the longitudinal and transverse dielectric functions including exciton correlations.

I. INTRODUCTION

With the recent advances in ultrashort pulse spectroscopy, it is now possible to observe in semiconductors electronic renormalizations induced by intense coherent laser beams. Such phenomena have been extensively studied in atomic systems.[1] In atomic physics, the analogy to effects previously considered has been often emphasized by using the same terminology. For example, in order to emphasize the analogy to the renormalizations in the photon vacuum, known as "Lamb shift," the high-field renormalizations are sometimes referred to as "lamp shift." Similarly, the analogy to dc field induced effects is reflected by such terms as "optical," "dynamical," or "ac Stark effect."

Clearly, pulses shorter than the relevant relaxation times allow to resolve similar coherent high-field phenomena in semiconductors as well, and this without damaging the samples. Recent experiments on quantum wells and bulk semiconductors under nonresonant below gap pulse excitation have revealed light-induced shifts of excitonic resonances and corresponding changes in the exciton oscillator strengths.[2–4] In these experiments the coherent electron-hole (e-h) pairs responsible for the excitonic optical Stark effect are generated only virtually and thus persist only as long as the pump beam is present. On the other hand, above gap excitation by ultrashort laser pulses has been used to generate high-density nonthermal e-h populations, and spectral hole burning in the interband optical absorption of semiconductors has been reported frequently. Except for a few cases, these investigations, performed in the context of hot-electron relaxation studies, have not paid much attention to the (initial) coherent time regime. This regime, depending on the excitation density and excess energy above the gap, can vary from a few hundred femtoseconds to a few femtoseconds.[5,6]

As already mentioned, these observations are similar to phenomena seen in atomic physics. However, the mutual interactions of the extended electronic excitations in semiconductors as well as their high mobility make the nature and description of these effects considerably different from the corresponding ones in atomic systems. Both the initially coherent interband absorption saturation and the excitonic optical Stark effect are manifestations of light-induced renormalizations which closely resemble those in superconductors and Bose-condensed systems, and can be described in the framework of one and the same theory. Their microscopic understanding is of great importance to fundamental solid-state theory as well as device applications in ultrafast optoelectronics. The purpose of the present paper is to give a comprehensive theory of these effects.

The formalism best suited to describe dense systems such as semiconductors is very different from that used in atomic physics, where the effects we consider have been observed first. Therefore, in order to establish a useful connection between the two descriptions, we will try to draw analogies when possible. Furthermore, in an effort to make the paper understandable to experimentalists, we will try to give intuitive interpretations of the concepts and the formalism we shall use, even if these are well known to the theorists. We shall also try to give plausible justifications of our rigorous results in order to help the impatient reader keeping track of the underlying physics.

The mobile and interacting electronic excitations in laser-excited semiconductors can be treated best by Green's-function techniques.[7] Frequently, the treatment can be simplified by using quasiequilibrium approximations.[7] The microscopic description of coherent high-field phenomena, however, requires nonequilibrium Green's-function techniques, as introduced by Keldysh[8] and Kadanoff and Baym.[9] Using this formalism, we derive in Sec. II the renormalized quasiparticle (e and h) spectra together with the coupled equations for the inversion and coherent polarization due to a strong monochromatic pump beam in the collisionless regime. We solve these equations analytically in Sec. III for the case of noninteracting e and h, where the problem reduces to that of a set of independent two-level systems. This section thus illustrates the application of the Green's-function technique to a well-known example. It shows that this very general formalism includes as a special case the treatment of light-matter interaction commonly employed in quantum electronics. We then include the influence of the Coulomb interactions within the Hartree-Fock approximation. We give analytical results for the nonlinear polarization induced by the pump field, both for the case of nonresonantly excited excitons and resonant interband excitation. In Sec. IV, we calculate the linear response of the renormalized system to weak perturbations, such as test beams or test charges. We give analytical results for the collective excitation spectrum as well as the longitudinal and transverse dielectric functions. The resulting absorption spectra are compared with available experimental data. Our theory is gauge invariant and fulfills the Ward identities exactly. Some of our results have already been reported in a recent paper.[10]

II. THE NONEQUILIBRIUM THEORY OF LASER-EXCITED SEMICONDUCTORS

Laser-excited semiconductors are examples of systems whose properties are strongly modified by external fields. The treatment of the optically created dense system of e and h requires the use of many-body nonequilibrium techniques.[7] Contrary to the situation in equilibrium systems, it is not sufficient to use only one type of Green's functions, e.g., the retarded one; this is because one has not only to calculate the renormalized quasiparticle spectra but also the corresponding nonequilibrium distribution functions. Keldysh[8] and Kadanoff and Baym[9] have shown how the relevant equations can be derived and how the various self-energies can be evaluated by a systematic diagram technique. Applications of this method to laser-excited semiconductors have been given, e.g., by Ivanov and Keldysh,[11] by Haug,[12] and by Schaefer and Treusch.[13] For an illustrative and detailed introduction into the technique, we refer to these papers as well as to two recent excellent review papers on quantum transport in metals.[14,15]

We consider homogeneously excited homogeneous semiconductors [either two dimensional (2D) or three dimensional (3D)], which we describe in a simple two-band model, with spin degeneracy only. The physics of coherently driven interband transitions in semiconductors is then very similar to that of superconductors[16–19] or Bose-condensed systems.[20–23] In the Green's-function formalism, the coherent polarization induced by a coherent laser field is determined by the off-diagonal interband Green's-function matrix element and corresponds to the anomalous Green's function (pair wave function) in a superconductor or the condensate wave function in a Bose condensate. However, the (e-h) "pairing" or "condensation" occurs in the particle-hole channel and it is not spontaneous, but externally enforced by the "symmetry breaking" pump field. With increasing pump intensity, i.e., e-h pair density, we recover the transition from real-space pairing (tightly bound excitons) to momentum-space pairing (weakly correlated, overlapping e-h Cooper pairs), discussed earlier in the context of spontaneous exciton condensation[24–28] and superconductivity.[29] Vice versa, we enrich the discussion of the latter phenomena by a consistent calculation of the collective excitation spectrum for arbitrary coupling, which interpolates smoothly between the Bogolubov-Beliaev theory of a weakly nonideal Bose gas[20,21] and the Anderson-Bogolubov theory of collective modes in superconductors.[17,18] Put another way, we extend the kinetic theory of weak coupling superconductors[19] to the strong coupling local regime, in which Fermi liquid theory breaks down. (In the theory of magnetism, this would correspond to the interpolation between itinerant and local moment behavior.)

Let $\psi_i(\mathbf{r}_1,t_1)\equiv\psi_i(1)$ be the field operators of electrons in the conduction ($i=1$) or valence ($i=2$) band. The 2×2 retarded Green's function is

$$G^r_{ij}(1,2)=-i\theta(t_1-t_2)\langle[\psi_i(1),\psi^\dagger_j(2)]_+\rangle\ . \tag{1}$$

The diagonal elements of $\hat{G}^r$ describe the propagation within the same band of an electron from point 2 to point 1. The off-diagonal elements describe the same propagation but with a transition from band j to band i. $\hat{G}^r$ determines the renormalized quasiparticle spectra and obeys a Dyson equation of the form

$$\begin{aligned}\hat{G}^r&=\hat{G}^r_0+\hat{G}^r_0\hat{\Sigma}^r\hat{G}^r\\&=\hat{G}^r_0+\hat{G}^r\hat{\Sigma}^r\hat{G}^r_0\ ,\end{aligned} \tag{2}$$

where integration over internal variables is implied. Spin indices are suppressed. G^r_0 is the unperturbed Green's function and $\hat{\Sigma}^r$ the retarded self-energy matrix. It describes the interactions with the total field felt by the particles which comprises the external field and the internal ("molecular") field. Inclusion of this latter, which accounts for the Coulomb coupling of the electronic excitations, is essential in semiconductors.

The interaction with the coherent monochromatic pump field, $E_p\exp(-i\omega_p t)+E_p^*\exp(i\omega_p t)$, will be treated in the rotating wave approximation, keeping resonant terms only. The rapid oscillations with frequency ω_p can be eliminated by working in the rotating frame. For simplicity, we do not treat explicitly the slower amplitude variations of the pulse; they can be included in a natural extension of the theory.

It is often advantageous to introduce relative ($\mathbf{r}=\mathbf{r}_1-\mathbf{r}_2$, $t=t_1-t_2$) and center [$R=(\mathbf{r}_1+\mathbf{r}_2)/2$, $T=(t_1+t_2)/2$] coordinates in space and time, as the microscopic and macroscopic scales, respectively. If the variations on both scales are sufficiently well separated, i.e., for slow variations on the macroscopic scale, one can Fourier transform Eq. (2) with respect to the relative coordinates $\mathbf{r}$ and t. In a spatially homogeneous situation, i.e., $\hat{G}^r$ independent of $\mathbf{R}$, one finds the adiabatic result ($\hbar=1$)

$$(2\omega+i0-\varepsilon^0_{i\mathbf{k}}-\varepsilon^0_{j\mathbf{k}})G^r_{ij}(\mathbf{k},\omega,T) = 2\delta_{ij}+\Sigma^r_{il}(\mathbf{k},\omega,T)G^r_{lj}(\mathbf{k},\omega,T) + G^r_{il}(\mathbf{k},\omega,T)\Sigma^r_{lj}(\mathbf{k},\omega,T) , \quad (3)$$

where

$$\varepsilon^0_{1\mathbf{k}}=(E_g-\omega_p)/2+k^2/(2m_e)$$

and

$$\varepsilon^0_{2\mathbf{k}}=-(E_g-\omega_p)/2-k^2/(2m_h)$$

are, respectively, the unperturbed conduction- and valence-band energies in the rotating frame. The solution of Eq. (3) is ($i\neq j$)

$$G^r_{ii}(\mathbf{k},\omega,T)=[\omega+i0-\varepsilon^0_{jk}-\Sigma^r_{jj}(\mathbf{k},\omega,T)]/D(\mathbf{k},\omega,T) \quad (4a)$$

and

$$G^r_{ij}(\mathbf{k},\omega,T)=\Sigma^r_{ij}(\mathbf{k},\omega,T)/D(\mathbf{k},\omega,T) , \quad (4b)$$

where

$$D(\mathbf{k},\omega,T)=[\omega+i0-\varepsilon^0_{1\mathbf{k}}-\Sigma^r_{11}(\mathbf{k},\omega,T)] \times[\omega+i0-\varepsilon^0_{2\mathbf{k}}-\Sigma^r_{22}(\mathbf{k},\omega,T)] -\Sigma^r_{12}(\mathbf{k},\omega,T)\Sigma^r_{21}(\mathbf{k},\omega,T) . \quad (4c)$$

In the present paper we consider two cases. The first is that of nonresonant coherent excitation well below the absorption edge. In this case no energy is deposited in the sample, the nonresonant field drives coherent valence charge fluctuations that can be viewed as "virtual e-h pairs." The coherence implies that these pairs experience no real collisions. In other words, the virtual transitions occur in a time shorter than $1/(E_g-\omega_p)$, which in turn must be shorter than any relaxation time in the medium. The other case is that of the initial transient that occurs for real transitions before the first collision. In the absence of both real *and* virtual (see below) collisions, the self-energy is instantaneous, i.e., independent of ω. We will denote it by $\hat{\Sigma}$, with matrix elements

$$\hat{\Sigma}(\mathbf{k},T)=\begin{pmatrix}\Sigma_{1\mathbf{k}} & -\Delta_{\mathbf{k}}\\ -\Delta^*_{\mathbf{k}} & \Sigma_{2\mathbf{k}}\end{pmatrix} . \quad (5)$$

The interband self-energy $\Delta_{\mathbf{k}}$ describes the e-h pairing due to the coherent pump field and the attractive e-h interaction, while the intraband self-energies $\Sigma_{1\mathbf{k}}$ and $\Sigma_{2\mathbf{k}}$ describe the renormalization of the paired e and h due to the e-e and h-h Coulomb repulsion, respectively. These two types of self-energies will be discussed in more detail later on.

In a nonequilibrium situation, in addition to the retarded Green's function, we have to evaluate the 2×2 distribution function

$$G^<_{ij}(1,2)=i\langle\psi^\dagger_j(2)\psi_i(1)\rangle . \quad (6)$$

$\hat{G}^<$ describes the correlation of electrons at points 1 and 2 and in bands i and j, and generalizes to quantum mechanical and interacting many-particle systems the usual concept of distribution function used in classical statistic mechanics. For $t_1=t_2$, $\hat{G}^<$ equals the reduced density matrix. The equation of motion for $\hat{G}^<$ is

$$\left[i\frac{\partial}{\partial T}-\varepsilon^0_{i1}+\varepsilon^0_{j2}\right]G^<_{ij}(1,2) =\Sigma^r_{il}(1,3)G^<_{lj}(3,2)-G^<_{il}(1,3)\Sigma^a_{lj}(3,2) -G^r_{il}(1,3)\Sigma^<_{lj}(3,2)+\Sigma^<_{il}(1,3)G^a_{lj}(3,2) , \quad (7)$$

where the superscript a denotes advanced functions and $\varepsilon_{l1}=\varepsilon_{l\mathbf{k}=-i\nabla_1}$. Equation (7) is the general transport equation of the system and formally exact. Under certain conditions it reduces to the well-known Boltzmann equation. Together with Eq. (2), it completely determines the electronic and optical properties of a laser-excited semiconductor.

In a spatially homogeneous situation and in the collisionless regime, $\hat{\Sigma}^<=0$ and $\hat{\Sigma}^r=\hat{\Sigma}^a=\hat{\Sigma}$, Eq. (7) can be reduced to an equation for the reduced density matrix

$$\left[i\frac{\partial}{\partial T}-\varepsilon^0_{i\mathbf{k}}+\varepsilon^0_{j\mathbf{k}}\right]n_{ij}(\mathbf{k},T) =\Sigma_{il}(\mathbf{k},T)n_{lj}(\mathbf{k},T)-n_{il}(\mathbf{k},T)\Sigma_{lj}(\mathbf{k},T) , \quad (8)$$

where

$$\hat{n}(\mathbf{k},T)=-i\int\frac{d\omega}{2\pi}\hat{G}^<(\mathbf{k},\omega,T) . \quad (9)$$

The matrix elements of $\hat{n}$ are

$$\hat{n}(\mathbf{k},T)=\begin{pmatrix}n_{1\mathbf{k}} & \psi_{\mathbf{k}}\\ \psi^*_{\mathbf{k}} & n_{2\mathbf{k}}\end{pmatrix} . \quad (10)$$

The interband matrix element $\psi_{\mathbf{k}}$ describes the polarization induced by the coherent pump field, while the intraband matrix elements $n_{1\mathbf{k}}$ and $n_{2\mathbf{k}}$ describe the nonequilibrium e and h distribution functions, respectively. Let us remark that Eq. (8) is nothing but the Heisenberg equation describing the evolution of the density matrix. It is necessary to derive Eq. (8) from Eq. (7) rather than introducing it directly because, as will be shown in Sec. IV, Eq. (8) *cannot* be used to describe the response of the system to external perturbations such as a test beam (a situation that we wish to treat). This is due to the long-range nature of the Coulomb interaction.

Equations (4), (5), (8), and (10) determine the coherent homogeneous "ground state" of a semiconductor, i.e., the condensate, driven by a strong monochromatic pump

beam, but without relaxation. Weak external perturbations, such as test beams or test charges, will excite the system out of its ground state, which is described by similar equations linearized in the perturbation. We will solve the equations in two steps, without and with Coulomb interactions. For noninteracting e and h the problem can be solved analytically, while in the presence of interactions only a few quantitative results can be obtained by hand.

III. THE COHERENT GROUND STATE

A. Noninteracting e-h pairs

For noninteracting e and h, the only self-energy in the problem is the radiative self-energy. It measures the strength of the interaction with the light field, i.e., $\Delta_{\mathbf{k}}^0=\mu E_p$, where μ is the interband dipole matrix element (assumed to be constant), and

$$\hat{\Sigma}_0(\mathbf{k},T)=\begin{pmatrix} 0 & -\mu E_p \\ -\mu^* E_p^* & 0 \end{pmatrix} . \tag{11}$$

Substituting Eq. (11) into Eq. (4), we find for the renormalized quasiparticle spectrum in the rotating frame (poles of the retarded Green's function)

$$\omega_{1,2\mathbf{k}}^0=\tfrac{1}{2}\{\varepsilon_{1\mathbf{k}}^0+\varepsilon_{2\mathbf{k}}^0\pm[(\varepsilon_{1\mathbf{k}}^0-\varepsilon_{2\mathbf{k}}^0)^2+4\,|\,\Delta_{\mathbf{k}}^0\,|^2]^{1/2}\} . \tag{12}$$

The renormalized e spectrum is

$$E_{1\mathbf{k}}^0=\begin{cases}\tfrac{1}{2}\{\varepsilon_{1\mathbf{k}}^0+\varepsilon_{2\mathbf{k}}^0+\omega_p+[(\varepsilon_{1\mathbf{k}}^0-\varepsilon_{2\mathbf{k}}^0)^2+4\,|\,\Delta_{\mathbf{k}}^0\,|^2]^{1/2}\}, & \varepsilon_{1\mathbf{k}}^0\geq\varepsilon_{2\mathbf{k}}^0 \\ \tfrac{1}{2}\{\varepsilon_{1\mathbf{k}}^0+\varepsilon_{2\mathbf{k}}^0+\omega_p-[(\varepsilon_{1\mathbf{k}}^0-\varepsilon_{2\mathbf{k}}^0)^2+4\,|\,\Delta_{\mathbf{k}}^0\,|^2]^{1/2}\}, & \varepsilon_{1\mathbf{k}}^0\leq\varepsilon_{2\mathbf{k}}^0 ,\end{cases} \tag{13a}$$

while the renormalized h spectrum is

$$E_{2\mathbf{k}}^0=\begin{cases}\tfrac{1}{2}\{\varepsilon_{1\mathbf{k}}^0+\varepsilon_{2\mathbf{k}}^0-\omega_p-[(\varepsilon_{1\mathbf{k}}^0-\varepsilon_{2\mathbf{k}}^0)^2+4\,|\,\Delta_{\mathbf{k}}^0\,|^2]^{1/2}\}, & \varepsilon_{1\mathbf{k}}^0\geq\varepsilon_{2\mathbf{k}}^0 \\ \tfrac{1}{2}\{\varepsilon_{1\mathbf{k}}^0+\varepsilon_{2\mathbf{k}}^0-\omega_p+[(\varepsilon_{1\mathbf{k}}^0-\varepsilon_{2\mathbf{k}}^0)^2+4\,|\,\Delta_{\mathbf{k}}^0\,|^2]^{1/2}\}, & \varepsilon_{1\mathbf{k}}^0\leq\varepsilon_{2\mathbf{k}}^0 .\end{cases} \tag{13b}$$

Figures 1(a) and 1(b) show $E_{1,2\mathbf{k}}^0$ for nonresonant ($\omega_p<E_g$) and resonant ($\omega_p>E_g$) excitation, respectively. In the case of nonresonant excitation, the states are blue shifted, the Stark shift decreasing with increasing detuning from the pump frequency. In the case of resonant excitation, gaps open at ω_p. For $\varepsilon_{1\mathbf{k}}^0=\varepsilon_{2\mathbf{k}}^0$, the state with N photons and the electron in the valence band is degenerate with the state with $N-1$ photons and the electron in the conduction band. The degeneracy is removed by the dipole interaction, which leads to the Stark splitting of the bands at ω_p. The magnitude of the light-induced gaps is given by $2\Delta_{\mathbf{k}}^0$, i.e., the Rabi frequency. Furthermore, the states with energy larger than $\omega_p+\Delta_{\mathbf{k}}^0$ are blue shifted, while those with energy smaller than $\omega_p-\Delta_{\mathbf{k}}^0$ are red shifted.

Substituting Eqs. (10) and (11) into Eq. (8), we find for the interband matrix element of the distribution function

$$\psi_{\mathbf{k}}^0=\frac{1-2n_{\mathbf{k}}^0}{\varepsilon_{1\mathbf{k}}^0-\varepsilon_{2\mathbf{k}}^0}\Delta_{\mathbf{k}}^0 \tag{14a}$$

$$=\frac{\mathrm{sgn}(\varepsilon_{1\mathbf{k}}^0-\varepsilon_{2\mathbf{k}}^0)}{[(\varepsilon_{1\mathbf{k}}^0-\varepsilon_{2\mathbf{k}}^0)^2+4\,|\,\Delta_{\mathbf{k}}^0\,|^2]^{1/2}}\Delta_{\mathbf{k}}^0 , \tag{14b}$$

and for the intraband matrix element $n_{\mathbf{k}}^0=n_{1\mathbf{k}}^0=1-n_{2\mathbf{k}}^0$

$$n_{\mathbf{k}}^0=\tfrac{1}{2}[1-(1-4\,|\,\psi_{\mathbf{k}}^0\,|^2)^{1/2}] \tag{15a}$$

$$=\frac{1}{2}\left[1-\frac{|\,\varepsilon_{1\mathbf{k}}^0-\varepsilon_{2\mathbf{k}}^0\,|}{[(\varepsilon_{1\mathbf{k}}^0-\varepsilon_{2\mathbf{k}}^0)^2+4\,|\,\Delta_{\mathbf{k}}^0\,|^2]^{1/2}}\right] . \tag{15b}$$

The second forms of Eqs. (14) and (15) can be derived from Eq. (15a). The e (h) population $n_{\mathbf{k}}^0$ is shown in Figs. 2(a) and 2(b) for nonresonant and resonant excitation, respectively. In the case of nonresonant excitation of virtual e and h, $n_{\mathbf{k}}^0$ is maximum at the gap and decreases smoothly with increasing detuning from the

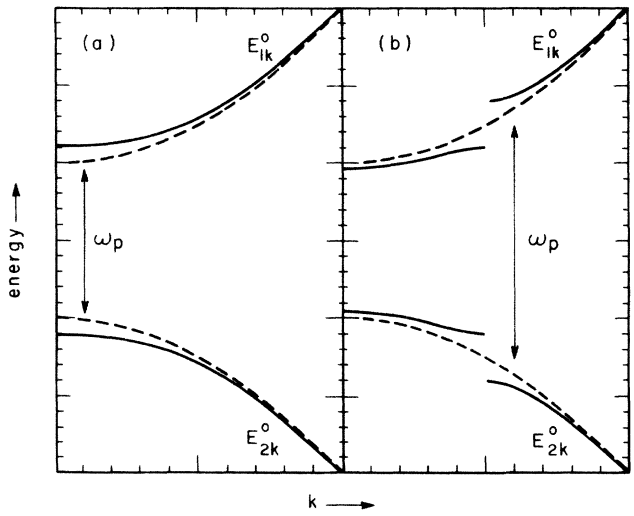

FIG. 1. Sketch of the renormalized conduction- (E_{1k}^0) and valence- (E_{2k}^0) band energies as a function of momentum k for $m_e=m_h$ and (a) nonresonant excitation of virtual e and h, $\omega_p<E_g$, (b) resonant excitation, $\omega_p>E_g$. The dashed lines show the unperturbed energies.

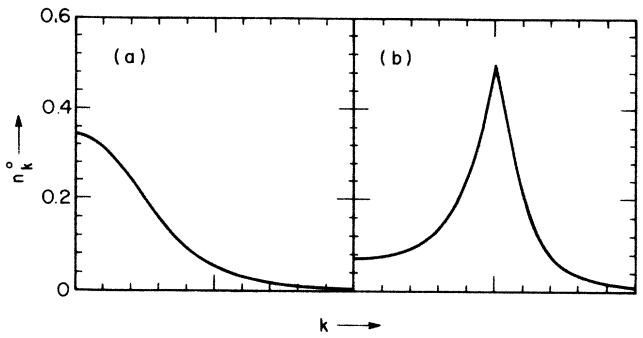

FIG. 2. Sketch of the fermion distribution function n_k^0 as a function of momentum k for (a) nonresonant excitation of virtual e and h, $\omega_p < E_g$, (b) resonant excitation, $\omega_p > E_g$.

pump frequency. In the case of resonant excitation, $n_{\mathbf{k}}^0$ peaks at ω_p and again decreases smoothly with increasing detuning. In the absence of relaxation, this is as expected. $n_{\mathbf{k}}^0 = \frac{1}{2}$ at ω_p corresponds to a complete bleaching of the pump beam absorption.

The polarization induced by the pump beam is

$$P_p = 2\mu^* \sum_{\mathbf{k}} \psi_{\mathbf{k}} \ . \tag{16}$$

Substituting Eq. (14) into Eq. (16), we obtain for the real nonlinear optical susceptibility, P_p/E_p,

$$\chi_p = 2|\mu|^2 \sum_{\mathbf{k}} \frac{1-2n_{\mathbf{k}}^0}{\varepsilon_{1\mathbf{k}}^0 - \varepsilon_{2\mathbf{k}}^0} = 2|\mu|^2 \sum_{\mathbf{k}} \frac{\mathrm{sgn}(\varepsilon_{1\mathbf{k}}^0 - \varepsilon_{2\mathbf{k}}^0)}{[(\varepsilon_{1\mathbf{k}}^0 - \varepsilon_{2\mathbf{k}}^0)^2 + 4|\Delta_{\mathbf{k}}^0|^2]^{1/2}} \ . \tag{17}$$

For small pump fields, $|E_p| \to 0$, this reduces to the usual linear response result, while for large pump fields, $|E_p| \to \infty$, χ_p saturates like the susceptibility of an inhomogeneously broadened two-level system. In the absence of Coulomb interactions, the saturation mechanism is phase-space filling (PSF) or "blocking," i.e., the states which are already occupied are no longer accessible in optical transitions, due to the Pauli exclusion principle. The third-order nonlinear optical susceptibility $\chi_p^{(3)}$ is

$$\chi_p^{(3)} = -4|\mu|^2 \sum_{\mathbf{k}} \frac{1}{(\varepsilon_{1\mathbf{k}}^0 - \varepsilon_{2\mathbf{k}}^0)^3} \ , \tag{18}$$

i.e., it shows resonance enhancement for $\omega_p \to E_g$.

The collisionless kinetic equation (8) has yet another solution. This is the quasiequilibrium solution that arises because the collision term vanishes in local equilibrium. This solution can also be obtained from the retarded Green's function $\hat{G}^r$

$$\hat{n}(\mathbf{k},T) = -\int \frac{d\omega}{2\pi} \frac{1}{e^{\beta\omega}+1} 2\,\mathrm{Im}\hat{G}^r(\mathbf{k},\omega,T) \ , \tag{19}$$

where β is the inverse temperature. At zero temperature one finds

$$n_{\mathbf{k}}^0 = \frac{1}{2}\left[1 - \frac{\varepsilon_{1\mathbf{k}}^0 - \varepsilon_{2\mathbf{k}}^0}{[(\varepsilon_{1\mathbf{k}}^0 - \varepsilon_{2\mathbf{k}}^0)^2 + 4|\Delta_{\mathbf{k}}^0|^2]^{1/2}}\right] \ , \tag{20}$$

which differs from Eq. (15) only by the fact that the numerator $\varepsilon_{1\mathbf{k}}^0 - \varepsilon_{2\mathbf{k}}^0 = E_g - \omega_p + k^2/(2m)$ (m is the reduced e-h mass, $m^{-1} = m_e^{-1} + m_h^{-1}$) can change sign for resonant excitation, $\omega_p > E_g$. In this case, $n_{\mathbf{k}}^0$ has the shape of a step function with a broadened edge. It corresponds to a radiation-broadened Fermi distribution at zero temperature and describes a completely relaxed degenerate e-h system with a gap at the quasichemical potential, the latter being identical to the pump frequency. This is the saturated state of a semiconductor. Starting with Galitskii *et al.*,[30] it has been studied by many authors, assuming that carrier-carrier and carrier-phonon collisions quickly drive the e and h towards thermal equilibrium.[12,31–34] Such a solution can, of course, *never* reduce to the linear response result. For nonresonant excitation, $\omega_p < E_g$, there is no difference to the nonequilibrium solution.

B. Interacting e-h pairs

So far, we have solved for the coherent ground state of noninteracting e and h, the results being equivalent to those obtained for an ensemble of independent two-level atoms.[1] This is as expected because optical transitions with different $\mathbf{k}$ decouple. Building on this simple case, we include now the coupling due to the Coulomb interactions, which we treat in the Hartree-Fock (HF) approximation.[10] Since we work in the electron picture, we have to subtract from $\varepsilon_{1\mathbf{k}}^0$ and $\varepsilon_{2\mathbf{k}}^0$ the interaction with the completely filled valence band (which is assumed to be already included in the parameters E_g, m_e, and m_h), so that in the following:

$$\varepsilon_{1\mathbf{k}}^0 = (E_g - \omega_p)/2 + k^2/(2m_e) - 2V_{q=0}\sum_{\mathbf{k}'} \ ,$$

and

$$\varepsilon_{2\mathbf{k}}^0 = -(E_g - \omega_p)/2 - k^2/(2m_h) - 2V_{q=0}\sum_{\mathbf{k}'} + \sum_{\mathbf{k}'} V_{\mathbf{k},\mathbf{k}'} \ ,$$

where $V_{\mathbf{k},\mathbf{k}'} = V_{\mathbf{q}=\mathbf{k}-\mathbf{k}'}$ is the Coulomb potential and $\sum_{\mathbf{k}}'$ runs over all valence-band states.

Note again, that "no relaxation" does *not* imply "no correlation," i.e., limitation to HF; we could allow for virtual collisions which would renormalize the Coulomb interaction, towards a Landau effective interaction in the weak coupling limit[19] and a T matrix in the strong coupling limit (see below).

Within the HF approximation, the contribution of the Coulomb interactions to the self-energy matrix is given by

$$\hat{\Sigma}_{\mathrm{HF}}(\mathbf{k},T) = 2V_{q=0}\sum_{\mathbf{k}'} \mathrm{tr}\hat{n}(\mathbf{k}',T) - \sum_{\mathbf{k}'} V_{\mathbf{k},\mathbf{k}'}\hat{n}(\mathbf{k}',T) \tag{21}$$

corresponding to renormalized conduction- and valence-band energies ($i,j=1,2$)

$$\varepsilon_{i\mathbf{k}}=\varepsilon^0_{i\mathbf{k}}+2V_{q=0}\sum_{j,\mathbf{k}'}n_{j\mathbf{k}'}-\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'}n_{i\mathbf{k}'}\ , \tag{22}$$

and a renormalized Rabi frequency

$$\Delta_{\mathbf{k}}=\Delta^0_{\mathbf{k}}+\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'}\psi_{\mathbf{k}'}\ . \tag{23}$$

These expressions show already what the effects of the Coulomb interactions are. They couple transitions with different $\mathbf{k}$, so that an e-h pair with given $\mathbf{k}$ does not only experience the external field alone; in addition, it also feels a significant internal field, the molecular field associated with e-h pairs created at $\mathbf{k}'$. At each $\mathbf{k}$, external and Coulomb fields combine to give an effective selfconsistent "local field." For small pump intensities, the local fields or "local-field corrections" dominate and transform the free e-h pairs into excitons (in the case of Frenkel excitons, we recover atomic behavior). This dilute limit corresponds to the usual description of excitonic effects, it demonstrates that the Coulomb coupling drastically changes the behavior of the interacting system as compared to the noninteracting case. For large pump intensities, the external field dominates. In this case, the governing processes are the individual band-to-band transitions. The Coulomb interaction only introduces a weak coupling compared to the external field which can be treated as a correction to the dominant processes. Thus we recover the transition from (atomic) real-space to collective momentum-space pairing.

In terms of the renormalized quantities $\varepsilon_{1\mathbf{k}}$, $\varepsilon_{2\mathbf{k}}$, and $\Delta_{\mathbf{k}}$, expressions (12)–(15) for the quasiparticle spectrum and distribution function remain unchanged. From Eq. (23), we obtain for the light-induced pair amplitude $\psi_{\mathbf{k}}$ the self-consistent equation[10]

$$(\varepsilon_{1\mathbf{k}}-\varepsilon_{2\mathbf{k}})\psi_{\mathbf{k}}=(n_{2\mathbf{k}}-n_{1\mathbf{k}})\left[\mu E_p+\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'}\psi_{\mathbf{k}'}\right], \tag{24a}$$

where

$$n_{1\mathbf{k}}=1-n_{2\mathbf{k}}=n_{\mathbf{k}}=\tfrac{1}{2}[1-(1-4\,|\,\psi_{\mathbf{k}}\,|^2)^{1/2}]\ . \tag{24b}$$

Without the nonlinear corrections, Eq. (24a) corresponds to an inhomogeneous Wannier equation in momentum space, driven by the pump field E_p. In this limit, it describes unperturbed excitons. The self-consistent nonlinear corrections describe the effects on the excitons of PSF and fermion exchange (E), through the factor $n_{2\mathbf{k}}-n_{1\mathbf{k}}$ and the exchange (F) self-energies contained in $\varepsilon_{1\mathbf{k}}$ and $\varepsilon_{2\mathbf{k}}$ [third term in Eq. (22)]. Note that in the homogeneous ground state the fermion Hartree (H) self-energies [second term in Eq. (22)] vanish, reflecting charge neutrality, i.e., the number of e equals the number of h.

In general, the nonlinear integral equation (24a) can only be solved numerically, e.g., by iteration or by integration for a pulsed pump field, and corresponding results will be given elsewhere. Here, we will only discuss its solution for nonresonant excitation of virtual e and h, $\omega_p<E_g-E_0$, where E_0 is the exciton binding energy, at moderate pump intensities. In this limit the excitons are still well defined and the nonlinear correction terms can be treated in perturbation theory. Together with the free-particle (i.e., external field dominated) results given above, this provides already a rather complete physical picture of a coherently driven semiconductor, covering both real and momentum space pairing.

For small pump intensities and $\omega_p<E_g-E_0$, i.e., in the linear-response regime, $\psi_{\mathbf{k}}\propto E_p$, we find from Eq. (24b) $n_{\mathbf{k}}\to|\psi_{\mathbf{k}}|^2\propto|E_p|^2$. The fermion distribution function is thus determined by the probability of finding the virtually excited e and h in bosonic exciton states. Substituting this result into Eq. (24a), we obtain the perturbed inhomogeneous Wannier equation[10]

$$\sum_{\mathbf{k}'}(H^0_{\mathbf{k},\mathbf{k}'}-\omega_p\delta_{\mathbf{k},\mathbf{k}'})\psi_{\mathbf{k}'}\to(1-2\,|\,\psi_{\mathbf{k}}\,|^2)\mu E_p-\sum_{\mathbf{k}'}\delta H^H_{\mathbf{k},\mathbf{k}'}\psi_{\mathbf{k}'}\ . \tag{25}$$

This equation deserves some comments. First, since

$$H^0_{\mathbf{k},\mathbf{k}'}=\left[E_g+\frac{k^2}{2m}\right]\delta_{\mathbf{k},\mathbf{k}'}-V_{\mathbf{k},\mathbf{k}'} \tag{26}$$

is the unperturbed e-h relative motion Hamiltonian, the left-hand side of Eq. (25) is simply the usual Wannier equation in momentum space written in the rotating frame. The first term on the right-hand side describes the coupling with the driving external field including the PSF reduction of the coupling constant expressed by the factor $(1-2\,|\,\psi_{\mathbf{k}}\,|^2)$. This PSF effect can be suitably reinterpreted as a correction due to the proper normalization of the exciton wave function in the presence of other e-h pairs. The second term on the right-hand side

$$\delta H^H_{\mathbf{k},\mathbf{k}'}=2n_{\mathbf{k}}V_{\mathbf{k},\mathbf{k}'}-2\sum_{\mathbf{k}''}V_{\mathbf{k},\mathbf{k}''}n_{\mathbf{k}''}\delta_{\mathbf{k},\mathbf{k}'}\to 2\,|\,\psi_{\mathbf{k}}\,|^2V_{\mathbf{k},\mathbf{k}'}-2\sum_{\mathbf{k}''}V_{\mathbf{k},\mathbf{k}''}\,|\,\psi_{\mathbf{k}''}\,|^2\delta_{\mathbf{k},\mathbf{k}'} \tag{27}$$

is the (boson Hartree) perturbation due to excitonic PSF [first term in Eq. (27)] and E [second term in Eq. (27)]. The non-Hermeticity of δH^H can, in principle, be removed by a simple transformation, which accounts for the PSF modification of the orthonormality relations.[7]

Equations (25)–(27) are of the form of the Gross-Pitaevskii equation for the order parameter of a weakly nonideal Bose condensate,[22,23] including a driving term and specialized to a spatially uniform situation. The order parameter is $\psi_{\mathbf{k}}$ and virtual exciton condensation occurs in *all* exciton states simultaneously, due to the nonresonant excitation (as discussed above, the pump frequency can be thought of as the quasichemical potential).

In order to clarify this point, we will now project Eq. (25) onto unperturbed exciton states. We will find that the self-energy and vertex corrections Eq. (27) combine simply to an effective exciton-exciton interaction. This transformation thus enables us to interpret the results rigorously derived so far in a more intuitive picture

where excitons are described as interacting "atomiclike particles." We can then make the connection with a number of more familiar concepts such as "hard-core repulsion" and "van der Waals attraction," the physics of which is more easy to feel.

Expanding $\psi_{\mathbf{k}}$ in terms of unperturbed exciton wave functions $\phi_{n\mathbf{k}}$

$$\psi_{\mathbf{k}}=\sum_n \psi_n \phi_{n\mathbf{k}} , \tag{28}$$

where

$$\sum_{\mathbf{k}'} H^0_{\mathbf{k},\mathbf{k}'}\phi_{n\mathbf{k}'}=E^0_n\phi_{n\mathbf{k}} , \tag{29}$$

we find that ψ_n, the projection of $\psi_{\mathbf{k}}$ on $\phi_{n\mathbf{k}}$, satisfies the equation

$$(E^0_n-\omega_p)\psi_n=\sum_{\mathbf{k}}(1-2\,|\,\psi_{\mathbf{k}}\,|^2)\phi^*_{n\mathbf{k}}\mu E_p-\sum_m \Sigma^H_{nm}\psi_m , \tag{30}$$

where Σ^H_{nm} is the Hartree exciton self-energy

$$\Sigma^H_{nm}=\sum_{\mathbf{k},\mathbf{k}'}\phi^*_{n\mathbf{k}}\delta H^H_{\mathbf{k},\mathbf{k}'}\phi_{m\mathbf{k}'} . \tag{31}$$

Using standard notation, Σ^H_{nm} can be expressed as[7]

$$\Sigma^H_{nm}\rightarrow\langle n\psi\,|\,I_x\,|\,m\psi\rangle , \tag{32}$$

where I_x is the exciton exchange interaction, i.e., $(V_t-V_s)/2$, where V_t and V_s are the exciton-exciton interactions in the triplet and singlet channels, respectively. The contribution of the direct exciton interaction I_d, i.e., $(V_t+V_s)/2$, to Eq. (30) vanishes, because of charge neutrality (this is, of course, only correct within the HF or Heitler-London approximation employed here). We will postpone to the end of this section the detailed physical interpretation of Eq. (30).

The nonlinear optical susceptibility is obtained from Eq. (16). Using Eqs. (29) and (30), we find in leading order[10]

$$\chi_p=2\sum_n\frac{f^H_n}{E^H_n-\omega_p} , \tag{33}$$

where

$$E^H_n=E^0_n+\Sigma^H_{nn} \tag{34}$$

are the renormalized transition energies and

$$f^H_n=|\,\mu\,|^2\sum_{\mathbf{k},\mathbf{k}'}\left[\phi_{n\mathbf{k}}(1-2\,|\,\psi_{\mathbf{k}'}\,|^2)\phi^*_{n\mathbf{k}'}-\sum_{\substack{m\\ m\neq n}}\frac{\phi_{n\mathbf{k}}\Sigma^H_{nm}\phi^*_{m\mathbf{k}'}+(n\leftrightarrow m)}{E^0_m-E^0_n}\right] \tag{35}$$

are the renormalized oscillator strengths. Here, it is understood that the linear response result

$$\psi_n=\sum_{\mathbf{k}}\frac{\phi^*_{n\mathbf{k}}}{E^0_n-\omega_p}\mu E_p=\frac{\phi^*_{n\mathbf{r}=0}}{E^0_n-\omega_p}\mu E_p \tag{36}$$

is substituted into Eqs. (34) and (35), so that the nonlinear corrections are of order $|\,E_p\,|^2$, i.e., linear in the density of virtually excited excitons

$$N=2\sum_{\mathbf{k}}|\,\psi_{\mathbf{k}}\,|^2=2\sum_n\frac{|\,\phi_{n\mathbf{r}=0}\,|^2\,|\,\mu E_p\,|^2}{(E^0_n-\omega_p)^2} . \tag{37}$$

For $|\,E_p\,|$, $N\rightarrow 0$, Eqs. (33)–(35) reduce to the exact linear response result, i.e., Elliott's formula.[35] For $|\,E_p\,|$, N finite, the dispersive optical nonlinearity is due to (i) a shift of the exciton energies as a result of exciton-exciton interactions, (ii) a corresponding exciton wave function renormalization [second term in Eq. (35)], and (iii) a PSF correction to the oscillator strength [first term in Eq. (35)]. Both (ii) and (iii) give rise to a negative $\chi^{(3)}_p$, i.e., a reduction of the exciton oscillator strength, because they both reduce the probability $|\,\phi_{n\mathbf{r}=0}\,|^2$ of finding the e and h in the same unit cell.

Equations (25)–(37) can as well be derived from an effective exciton Hamiltonian, with two basic ingredients, (i) an anharmonic exciton-photon interaction and (ii) an anharmonic exciton-exciton interaction.[36] The former is obtained from the linear exciton-photon interaction by projecting out the states which are already occupied, i.e., by preventing "double occupancy;" in the ground state, it gives rise to the PSF correction to the oscillator strengths. The latter is the analogue of "superexchange" in the theory of magnetism and, in the ground state, gives rise to the exciton self-energy Σ^H_{nm}, which, in general, would be rigorously determined by the exciton T matrix, rather than by the bare interaction I, as in our approach. Besides the short-range hard-core repulsion of excitons already contained in Eq. (32) (which, as evident from our discussion, results from the underlying Fermi statistics), the full T matrix would also describe their van der Waals attraction, i.e., mutual exciton polarization formally determined by "intermediate-state interactions." The problem with these as well as "final-state interactions" is that the T matrix in the singlet channel diverges, signaling the instability of the system towards biexciton formation. The divergence can be removed, however, by allowing for coexistence of virtual particle (excitons) and molecule (biexcitons) condensates, following our own and others earlier work on attractive Bose systems.[7,37,38] This would then also allow for a rigorous description of coherent two-photon generation of biexcitons, which so far has been treated rather naively.[7,39]

To conclude this section, let us note again that the above discussion is limited to low intensities, where excitons are well defined and the problem is that of a weakly nonideal virtual exciton gas. With increasing intensity, the e-h pairs spread out and overlap, and saturation sets

in. The latter is nothing but the quantum saturation of the exciton states. At high intensities, the system is best thought of in terms of decoupled interband transitions with only quantitative modifications due to the Coulomb interactions.

IV. COLLECTIVE EXCITATIONS AND THE LINEAR RESPONSE TO WEAK PERTURBATIONS

The changes of the system properties due to the action of the coherent pump field can, e.g., be determined by measuring the linear response to weak perturbations, such as test beams or test charges, as characterized by respective dielectric functions. Theoretically, the latter can either be obtained from the Kubo formula for the corresponding correlation functions or directly from the transport equation. Both methods are equivalent and yield exactly the same answer. Here, we will choose the second method, i.e., the solution of Eq. (7) linearized in the perturbation. Note that because of the long-range nature of the Coulomb interactions, it is *not* possible to linearize directly Eq. (8), even if the perturbation conserves momentum.

Any weak external perturbation of the system can be expressed as a small change $\delta\hat{\varepsilon}_{\rm ext}$ of the ground-state energy matrix

$$\hat{\varepsilon}(\mathbf{k},T)=\begin{bmatrix}\varepsilon^0_{1\mathbf{k}} & 0\\ 0 & \varepsilon^0_{2\mathbf{k}}\end{bmatrix}+\hat{\Sigma}_0(\mathbf{k},T)+\hat{\Sigma}_{\rm HF}(\mathbf{k},T)\ , \tag{38}$$

which enters the Heisenberg equation (8)

$$i\frac{\partial}{\partial T}\hat{n}(\mathbf{k},T)=[\hat{\varepsilon}(\mathbf{k},T),\hat{n}(\mathbf{k},T)]_-\ . \tag{39}$$

In addition to the direct change $\delta\hat{\varepsilon}_{\rm ext}$ of the ground-state energy matrix, there will be further changes through the "reaction" of the system, as discussed previously. Thus in order to avoid unphysical results, the total change of the ground-state energy matrix has to be evaluated self-consistently. First, if $\delta\hat{\varepsilon}_{\rm ext}$ does not conserve momentum, i.e., $\delta\hat{\varepsilon}_{\rm ext}=\delta\hat{\varepsilon}_{\rm ext}(\mathbf{k},\mathbf{q},T)$, it induces through Eq. (7) small fluctuations

$$\delta\hat{n}(\mathbf{k},\mathbf{q},T)=-i\int d^dR\int\frac{d\omega}{2\pi}e^{-i\mathbf{q}\cdot\mathbf{R}}\delta\hat{G}^<(\mathbf{k},\mathbf{R},\omega,T) \tag{40}$$

about the homogeneous ground-state distribution $\hat{n}$. In turn, through Eq. (21), these fluctuations give rise to local-field corrections

$$\delta\hat{\Sigma}_{\rm HF}(\mathbf{k},\mathbf{q},T)=2V_q\sum_{\mathbf{k}'}{\rm tr}\delta\hat{n}(\mathbf{k}',\mathbf{q},T)-\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'}\delta\hat{n}(\mathbf{k},'\mathbf{q},T)\ , \tag{41}$$

so that the total local field to which the system responds is

$$\delta\hat{\varepsilon}(\mathbf{k},\mathbf{q},T)=\delta\hat{\varepsilon}_{\rm ext}(\mathbf{k},\mathbf{q},T)+\delta\hat{\Sigma}_{\rm HF}(\mathbf{k},\mathbf{q},T)\ . \tag{42}$$

Finally, the linearization of Eq. (7) yields

$$i\frac{\partial}{\partial T}\delta\hat{n}(\mathbf{k},\mathbf{q},T)=\hat{\varepsilon}\left[\mathbf{k}-\frac{\mathbf{q}}{2},T\right]\delta\hat{n}(\mathbf{k},\mathbf{q},T)-\delta\hat{n}(\mathbf{k},\mathbf{q},T)\hat{\varepsilon}\left[\mathbf{k}+\frac{\mathbf{q}}{2},T\right]+\delta\hat{\varepsilon}(\mathbf{k},\mathbf{q},T)\hat{n}\left[\mathbf{k}+\frac{\mathbf{q}}{2},T\right]-\hat{n}\left[\mathbf{k}-\frac{\mathbf{q}}{2},T\right]\delta\hat{\varepsilon}(\mathbf{k},\mathbf{q},T)\ . \tag{43}$$

This set of coupled equations for the induced charge density and pair fluctuations is equivalent to the summation of bubble [first (H) term in Eq. (41)] and ladder [second (F) term in Eq. (41)] diagrams in Nambu space. As evident from Eqs. (38) and (41)–(43), self-energy and vertex corrections are treated on equal footing, so that the Ward identities are fulfilled exactly and our theory is fully gauge invariant.

The Hartree contribution to the local-field, Eq. (42), describes the screening of the external perturbation by induced charge density fluctuations. Its importance in a similar context was first pointed out by Anderson who showed that in superconductors it pushes the order parameter phase mode, the Anderson-Bogolubov mode, up to the plasma frequency.[17] Here, we are dealing with a neutral system so that low-lying collective excitations do indeed exist.

The induced charge density fluctuations can be integrated out exactly. From Eq. (8) or (39) one finds the conservation laws

$$i\frac{\partial}{\partial T}\{4n_{12}(\mathbf{k},T)n_{21}(\mathbf{k},T)+[n_{11}(\mathbf{k},T)-n_{22}(\mathbf{k},T)]^2\}=0\ , \tag{44a}$$

$$i\frac{\partial}{\partial T}[n_{11}(\mathbf{k},T)+n_{22}(\mathbf{k},T)]=0\ , \tag{44b}$$

the solution of which is still formally given by Eq. (15) with, however, renormalized parameters, i.e., Eq. (24b). Therefore, using $n_{\mathbf{k}}=n_{1\mathbf{k}}=1-n_{2\mathbf{k}}$, we find

$$\delta n_{11}(\mathbf{k},T)=-\delta n_{22}(\mathbf{k},T)=\frac{\psi^*_k\delta n_{12}(\mathbf{k},T)+\psi_{\mathbf{k}}\delta n_{21}(\mathbf{k},T)}{1-2n_{\mathbf{k}}}\ . \tag{45}$$

The extension to finite $\mathbf{q}$ of this relation is[10]

$$\delta n_{11}(\mathbf{k},\mathbf{q},T)=\frac{\psi^*_{\mathbf{k}-\mathbf{q}/2}\,\delta n_{12}(\mathbf{k},\mathbf{q},T)+\psi_{\mathbf{k}+\mathbf{q}/2}\,\delta n_{21}(\mathbf{k},\mathbf{q},T)}{1-n_{\mathbf{k}-\mathbf{q}/2}-n_{\mathbf{k}+\mathbf{q}/2}}\,, \tag{46a}$$

$$\delta n_{22}(\mathbf{k},\mathbf{q},T)=-\frac{\psi^*_{\mathbf{k}+\mathbf{q}/2}\,\delta n_{12}(\mathbf{k},\mathbf{q},T)+\psi_{\mathbf{k}-\mathbf{q}/2}\,\delta n_{21}(\mathbf{k},\mathbf{q},T)}{1-n_{\mathbf{k}-\mathbf{q}/2}-n_{\mathbf{k}+\mathbf{q}/2}}\,, \tag{46b}$$

which automatically takes care of two of the four equations (43). The remaining two are just the complex conjugate of each other.

In the rotating frame, a weak test beam, $E_t\exp(-i\omega_t t)+E_t^*\exp(i\omega_t t)$, gives rise to a perturbation

$$\delta\hat{\varepsilon}_{\rm ext}(\mathbf{k},T)=\begin{bmatrix}0 & -\mu E_t e^{-i\Delta\omega T}\\ -\mu^* E_t^* e^{i\Delta\omega T} & 0\end{bmatrix}\,, \tag{47}$$

where $\Delta\omega=\omega_t-\omega_p$. In the following, we will calculate the polarization induced by such a perturbation in the presence of the pump field E_p. Again, we shall treat both cases of interacting and noninteracting e and h.

Substituting Eqs. (46) and (47) into Eq. (43), we are left with an equation for the induced pair fluctuations, which in the long-wavelength limit $q\to 0$ reads [$\delta\psi_{\mathbf{k}}=\lim_{q\to 0}\delta n^*_{21}(\mathbf{k},\mathbf{q},T)=\lim_{q\to 0}\delta n_{12}(\mathbf{k},\mathbf{q},T)$]

$$\begin{aligned}i\frac{\partial}{\partial T}\delta\psi_{\mathbf{k}}=&(\varepsilon_{1\mathbf{k}}-\varepsilon_{2\mathbf{k}})\delta\psi_{\mathbf{k}}-(n_{2\mathbf{k}}-n_{1\mathbf{k}})\left[\mu E_t e^{-i\Delta\omega T}+\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'}\delta\psi_{\mathbf{k}'}\right]\\&+2\Delta_{\mathbf{k}}\frac{\psi^*_{\mathbf{k}}\delta\psi_{\mathbf{k}}+\psi_{\mathbf{k}}\delta\psi^*_{\mathbf{k}}}{n_{2\mathbf{k}}-n_{1\mathbf{k}}}-2\psi_{\mathbf{k}}\sum_{\mathbf{k}'}V_{\mathbf{k},\mathbf{k}'}\frac{\psi^*_{\mathbf{k}'}\delta\psi_{\mathbf{k}'}+\psi_{\mathbf{k}'}\delta\psi^*_{\mathbf{k}'}}{n_{2\mathbf{k}'}-n_{1\mathbf{k}'}}\\&-\lim_{q\to 0}\left[\mathbf{q}\cdot\frac{\partial\psi_{\mathbf{k}}}{\partial\mathbf{k}}\right]2V_q\sum_{\mathbf{k}'}\frac{\left[\mathbf{q}\cdot\frac{\partial\psi_{\mathbf{k}'}}{\partial\mathbf{k}'}\right]\delta\psi^*_{\mathbf{k}'}-\left[\mathbf{q}\cdot\frac{\partial\psi^*_{\mathbf{k}'}}{\partial\mathbf{k}'}\right]\delta\psi_{\mathbf{k}'}}{n_{2\mathbf{k}'}-n_{1\mathbf{k}'}}\,.\end{aligned} \tag{48}$$

Like Eq. (24a), this equation is of the form of an inhomogeneous Wannier equation, linearly driven by the test field E_t but nonlinear in the pump field E_p. The physical meaning of the various nonlinear terms becomes obvious if we eliminate the time dependence of $\delta\psi_{\mathbf{k}}$. Using the Ansatz

$$\delta\psi_{\mathbf{k}}=\delta\psi^+_{\mathbf{k}}e^{-i\Delta\omega T}+(\delta\psi^-_{\mathbf{k}})^*e^{i\Delta\omega T}\,, \tag{49}$$

we find the coupled Wannier equations

$$\sum_{\mathbf{k}'}[(\Delta\omega+i0+\omega_p)\delta_{\mathbf{k},\mathbf{k}'}-H^0_{\mathbf{k},\mathbf{k}'}]\delta\psi^+_{\mathbf{k}'}=-(1-2n_{\mathbf{k}})\mu E_t+\sum_{\mathbf{k}'}[(\delta H^H_{\mathbf{k},\mathbf{k}'}+\delta H^F_{\mathbf{k},\mathbf{k}'})\delta\psi^+_{\mathbf{k}'}+(\delta H^B_{\mathbf{k},\mathbf{k}'})\delta\psi^-_{\mathbf{k}'}]\,, \tag{50a}$$

$$\sum_{\mathbf{k}'}[(\Delta\omega+i0-\omega_p)\delta_{\mathbf{k},\mathbf{k}'}+H^0_{\mathbf{k},\mathbf{k}'}]\delta\psi^-_{\mathbf{k}'}=-\sum_{\mathbf{k}'}[(\delta H^H_{\mathbf{k},\mathbf{k}'}+\delta H^F_{\mathbf{k},\mathbf{k}'})^*\delta\psi^-_{\mathbf{k}'}+(\delta H^B_{\mathbf{k},\mathbf{k}'})^*\delta\psi^+_{\mathbf{k}'}]\,, \tag{50b}$$

where the unperturbed e-h relative motion Hamiltonian H^0 and the nonlinear boson Hartree correction δH^H are defined in Eqs. (26) and (27). The two other terms δH^F and δH^B are nonlinear boson Fock and Bogolubov corrections which read, respectively,

$$\delta H^F_{\mathbf{k},\mathbf{k}'}=\left[2\Delta^0_{\mathbf{k}}\psi^*_{\mathbf{k}}\delta_{\mathbf{k},\mathbf{k}'}+2\psi^*_{\mathbf{k}}\sum_{\mathbf{k}''}V_{\mathbf{k},\mathbf{k}''}\psi_{\mathbf{k}''}\delta_{\mathbf{k},\mathbf{k}'}-2\psi_{\mathbf{k}}V_{\mathbf{k},\mathbf{k}'}\psi^*_{\mathbf{k}'}+\lim_{q\to 0}\left[\mathbf{q}\cdot\frac{\partial\psi_{\mathbf{k}}}{\partial\mathbf{k}}\right]2V_q\left[\mathbf{q}\cdot\frac{\partial\psi^*_{\mathbf{k}'}}{\partial\mathbf{k}'}\right]\right]\Big/(1-2n_{\mathbf{k}'})\,, \tag{51}$$

and

$$\delta H^B_{\mathbf{k},\mathbf{k}'}=\left[2\Delta^0_{\mathbf{k}}\psi_{\mathbf{k}}\delta_{\mathbf{k},\mathbf{k}'}+2\psi_{\mathbf{k}}\sum_{\mathbf{k}''}V_{\mathbf{k},\mathbf{k}''}\psi_{\mathbf{k}''}\delta_{\mathbf{k},\mathbf{k}'}-2\psi_{\mathbf{k}}V_{\mathbf{k},\mathbf{k}'}\psi_{\mathbf{k}'}-\lim_{q\to 0}\left[q\cdot\frac{\partial\psi_{\mathbf{k}}}{\partial\mathbf{k}}\right]2V_q\left[q\cdot\frac{\partial\psi_{\mathbf{k}'}}{\partial\mathbf{k}'}\right]\right]\Big/(1-2n_{\mathbf{k}'})\,. \tag{52}$$

Equations (50) show that the structure of Eq. (48) is indeed very simple. Besides the PSF modification of the driving term linear in the test field, there are two elementary nonlinear processes, which determine the linear response to a weak test beam; namely, (i) e-h-pair renormalization described by the H and F terms, and (ii) e-h pair-pair creation and annihilation described by the B term. The H and F terms do not couple $\delta\psi^+_{\mathbf{k}}$ and $\delta\psi^-_{\mathbf{k}}$ whereas B does. The former process (H and F), renormalization of e-h pairs excited out of the coherent ground state (condensate), should not be confused with the renormalization of the condensate e-h pairs themselves which was treated in Sec. III. It is both due to the anharmonic interaction with pump photons [first term in Eq. (51)] and with other e-h pairs in the condensate [Eq. (27) and remaining terms in Eq. (51)]. In the latter process, creation and annihilation of pairs of excited e-h pairs, two pump photons, and thus two e-h pairs

out of the condensate are destroyed and two renormalized excited e-h pairs are created, one of which eventually transforms into a test photon, and vice versa.[1] Again, as evident from Eq. (52), both anharmonic interactions contribute to this nonlinear process, the "depletion of the condensate" stimulated by the presence of test photons, which necessarily leads to optical gain, even though there is *no* population inversion.

The polarization induced by the test beam is

$$P_t = 2\mu^* \sum_{\mathbf{k}} \delta\psi_{\mathbf{k}}^+ \ . \tag{53}$$

A. Noninteracting e-h pairs

For noninteracting e and h, $V_q = 0$, the only anharmonic interaction in the problem is that with the pump photons, and Eq. (50) can be solved analytically. Substituting the resulting expression for $\delta\psi_{\mathbf{k}}^+$ into Eq. (53), we obtain for the optical susceptibility experienced by the test beam, P_t/E_t,

$$\chi_t = 2\,|\,\mu\,|^2 \sum_{\mathbf{k}} \left[\frac{(1-n_{\mathbf{k}}^0)^2}{E_{1\mathbf{k}}^0 - E_{2\mathbf{k}}^0 - \omega_t - i0} - \frac{(n_{\mathbf{k}}^0)^2}{2\omega_p - E_{1\mathbf{k}}^0 + E_{2\mathbf{k}}^0 - \omega_t - i0} \right] , \tag{54}$$

where the ground-state distribution function, $n_{\mathbf{k}}^0$, and the renormalized conduction- and valence-band energies, $E_{1\mathbf{k}}^0$ and $E_{2\mathbf{k}}^0$, are defined in Eqs. (15) and (13), respectively. This result is identical to that derived for the case of two-level atoms.[1] Again, it demonstrates that the simple case of noninteracting "atomic" systems is included in our general formalism.

The integrand in Eq. (54) exhibits poles at $\Delta\omega = \pm\omega_{\mathbf{k}}^0$

$$\omega_{\mathbf{k}}^0 = \omega_{1\mathbf{k}}^0 - \omega_{2\mathbf{k}}^0 = [(\varepsilon_{1\mathbf{k}}^0 - \varepsilon_{2\mathbf{k}}^0)^2 + 4\,|\,\Delta_{\mathbf{k}}^0\,|^2]^{1/2} , \tag{55}$$

$\omega_{\mathbf{k}}^0$ are the frequencies of collective excitations above the condensate, i.e., the eigenmodes of Eq. (50). In the case of nonresonant excitation of virtual e and h, $\omega_p < E_g$, the excitation spectrum has a gap,

$$\min_{\mathbf{k}} \omega_{\mathbf{k}}^0 = \omega_{k=0}^0 = [(E_g - \omega_p)^2 + 4\,|\,\mu E_p\,|^2]^{1/2} \rightarrow E_g - \omega_p + \frac{2\,|\,\mu E_p\,|^2}{E_g - \omega_p} \tag{56}$$

in leading order in the pump intensity, which is just the minimum energy required to create real Stark shifted e-h pairs. As evident from Fig. 1(a) and shown in Fig. 3(a), optical absorption of test photons occurs at $\omega_t = \omega_p + \omega_{\mathbf{k}}^0 = E_{1\mathbf{k}}^0 - E_{2\mathbf{k}}^0$, i.e., for $\omega_t > \omega_p + \omega_{k=0}^0 > E_g$, and corresponds to the creation of renormalized excited e-h pairs and destruction of a test photon [first term in Eq. (54)]. Optical gain occurs at a photon energy symmetric about ω_p, at $\omega_t = \omega_p - \omega_{\mathbf{k}}^0$, i.e., for $\omega_t < \omega_p - \omega_{k=0}^0 < 2\omega_p - E_g$, and corresponds to the simultaneous emission of a test photon and a collective excitation, or, with $\omega_p - \omega_{\mathbf{k}}^0 = 2\omega_p - (\omega_p + \omega_{\mathbf{k}}^0) = 2\omega_p - E_{1\mathbf{k}}^0 + E_{2\mathbf{k}}^0$, to the depletion of the condensate; two pump photons are destroyed while a test photon and renormalized excited e-h pair are created [second term in Eq. (54)]. Between the regions of absorption and gain, there is a spectral region of width $2\min_{\mathbf{k}} \omega_{\mathbf{k}}^0 = 2\omega_{k=0}^0 > 2(E_g - \omega_p)$, in which the crystal is transparent.

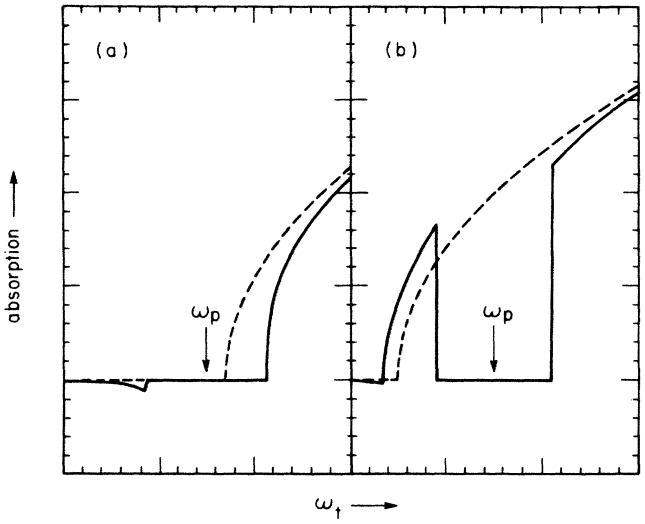

FIG. 3. Sketch of the test beam absorption as a function of frequency ω_t for (a) nonresonant excitation of virtual e and h, $\omega_p < E_g$, (b) resonant excitation, $\omega_p > E_g$. The dashed lines show the unperturbed absorption.

In the case of resonant excitation, $\omega_p > E_g$, the excitation spectrum has a gap

$$\min_{\mathbf{k}} \omega_{\mathbf{k}}^0 = 2\,|\,\mu E_p\,| \ , \tag{57}$$

which yields a spectral hole of width $4\,|\,\mu E_p\,|$ about ω_p, in which the crystal is transparent. As evident from Fig. 1(b) and shown in Fig. 3(b), optical absorption into renormalized excited e-h pair states occurs both below, $\omega_p - \omega_{k=0}^0 < \omega_t < \omega_p - 2\,|\,\mu E_p\,|$, and above, $\omega_t > \omega_p + 2\,|\,\mu E_p\,|$, this region, with spectral weight $(1-n_{\mathbf{k}}^0)^2 > \frac{1}{4}$. Optical gain occurs for $\omega_p + 2\,|\,\mu E_p\,| < \omega_t < \omega_p + \omega_{k=0}^0$ and $\omega_t < \omega_p - 2\,|\,\mu E_p\,|$, with spectral weight $(n_{\mathbf{k}}^0)^2$, i.e., regions of absorption and gain overlap for $\omega_p - \omega_{k=0}^0 < \omega_t < \omega_p - 2\,|\,\mu E_p\,|$ and $\omega_p + 2\,|\,\mu E_p\,| < \omega_t < \omega_p + \omega_{k=0}^0$, whereby, with $1 - n_{\mathbf{k}}^0 > n_{\mathbf{k}}^0$, the absorption always dominates.

For small pump fields, $|\,E_p\,| \rightarrow 0$, Eq. (54) reduces to the exact linear response result. The Stark shift occurs in first order in the pump intensity, while the optical gain is at least quadratic in the pump intensity, because two pump photons are required. The results for resonant excitation should be contrasted with those for the saturated state of a semiconductor mentioned in Sec. III. In the latter case, real e-h pairs exist everywhere below ω_p, so that (i) additional absorption and emission processes are possible and (ii) the spectral weights are changed. This results in optical gain below, $\omega_t < \omega_p - 2\,|\,\mu E_p\,|$, and optical absorption above, $\omega_t > \omega_p + 2\,|\,\mu E_p\,|$, the spectral hole.[33]

B. Interacting e-h pairs

For interacting e and h, Eq. (50) can only be solved numerically. Therefore, we limit ourselves again to nonresonant excitation of virtual e and h, $\omega_p < E_g - E_0$, and small pump intensities, where we can expand the pair wave function about unperturbed exciton states. As in Sec. III, this amounts to replacing $n_{\mathbf{k}}$ by $|\psi_{\mathbf{k}}|^2$ in Eqs. (27) and (50a) and, moreover, neglecting $n_{\mathbf{k}}$ in the denominator of expressions (51) and (52). The resulting perturbations δH^H, δH^F, and δH^B are then quadratic in the condensate wave function $\psi_{\mathbf{k}}$, i.e., the pump field E_p, and we recover exactly the $q=0$ Bogolubov-Beliaev equations for the excitation spectrum of a weakly nonideal Bose gas,[20,21] including a driving term.

Expanding $\delta\psi_{\mathbf{k}}^+(\delta\psi_{\mathbf{k}}^-)$ in terms of unperturbed exciton wave functions $\phi_{n\mathbf{k}}(\phi_{n\mathbf{k}}^*)$, Eq. (29),

$$\delta\psi_{\mathbf{k}}^+ = \sum_n \delta\psi_n^+ \phi_{n\mathbf{k}} , \tag{58a}$$

$$\delta\psi_{\mathbf{k}}^- = \sum_n \delta\psi_n^- \phi_{n\mathbf{k}}^* , \tag{58b}$$

we find that the projections satisfy the relations[10]

$$(\Delta\omega + i0 + \omega_p - E_n^0)\delta\psi_n^+ \rightarrow -\sum_{\mathbf{k}} (1-2|\psi_{\mathbf{k}}|^2)\phi_{n\mathbf{k}}^* \mu E_t + \sum_m [(\Sigma_{nm}^H + \Sigma_{nm}^F)\delta\psi_m^+ + (\Sigma_{nm}^B)\delta\psi_m^-] , \tag{59a}$$

$$(\Delta\omega + i0 - \omega_p + E_n^0)\delta\psi_n^- \rightarrow -\sum_m [(\Sigma_{nm}^H + \Sigma_{nm}^F)^* \delta\psi_m^- + (\Sigma_{nm}^B)^* \delta\psi_m^+] . \tag{59b}$$

In these equations, Σ_{nm}^H is the Hartree exciton self-energy defined in Eqs. (31) and (32). Σ_{nm}^F and Σ_{nm}^B are Fock and Bogolubov exciton self-energies which read, respectively,

$$\Sigma_{nm}^F = \sum_{\mathbf{k},\mathbf{k}'} \phi_{n\mathbf{k}}^* \delta H_{\mathbf{k},\mathbf{k}'}^F \phi_{m\mathbf{k}'} \rightarrow \Pi_{nm}^F + 2\langle n\psi | I_d | \psi m\rangle + \langle n\psi | I_x | \psi_m\rangle \tag{60}$$

and

$$\Sigma_{nm}^B = \sum_{\mathbf{k},\mathbf{k}'} \phi_{n\mathbf{k}}^* \delta H_{\mathbf{k},\mathbf{k}'}^B \phi_{m\mathbf{k}'}^* \rightarrow \Pi_{nm}^B + 2\langle nm | I_d | \psi\psi\rangle + \langle nm | I_x | \psi\psi\rangle . \tag{61}$$

In Eqs. (60) and (61), in order to make clear the physical origin of the various interaction terms, we have explicitly separated the contributions due to the anharmonic exciton-exciton interaction from the anharmonic exciton-photon interaction, Π_{nm}^F and Π_{nm}^B,

$$\Pi_{nm}^F = 2\mu E_p \sum_{\mathbf{k}} \phi_{n\mathbf{k}}^* \psi_{\mathbf{k}}^* \phi_{m\mathbf{k}} \tag{62}$$

and

$$\Pi_{nm}^B = 2\mu E_p \sum_{\mathbf{k}} \phi_{n\mathbf{k}}^* \psi_{\mathbf{k}} \phi_{m\mathbf{k}}^* . \tag{63}$$

The direct exciton-exciton interaction I_d in Eqs. (60) and (61) appears as a result of screening.

In leading order in the pump intensity, exciton pair creation, and annihilation, as described by the Bogolubov self-energies, can be neglected, so that the only nonlinear processes left are PSF and renormalization of excited e-h pairs. The optical susceptibility experienced by the test beam is[10]

$$\chi_t = 2\sum_n \frac{f_n^{\mathrm{HF}}}{E_n^{\mathrm{HF}} - \omega_t - i0} , \tag{64}$$

where

$$E_n^{\mathrm{HF}} = E_n^0 + \Sigma_{nn}^H + \Sigma_{nn}^F \tag{65}$$

are the renormalized transition energies and

$$f_n^{\mathrm{HF}} = |\mu|^2 \sum_{\mathbf{k},\mathbf{k}'} \left[\phi_{n\mathbf{k}}(1-2|\psi_{\mathbf{k}'}|^2)\phi_{n\mathbf{k}'}^* - \sum_{\substack{m \\ m\neq n}} \frac{\phi_{n\mathbf{k}}(\Sigma_{nm}^H + \Sigma_{nm}^F)\phi_{m\mathbf{k}'}^* + (n \leftrightarrow m)}{E_m^0 - E_n^0} \right] \tag{66}$$

the renormalized oscillator strengths. Here, it is again understood that the linear response result Eq. (36) is substituted into Eqs. (65) and (66), so that the nonlinear corrections are of order $|E_p|^2$, i.e., linear in the density N of virtually excited excitons, Eq. (37).

The comparison of χ_t with the optical susceptibility χ_p experienced by the pump beam, Eqs. (33)–(35), reveals the different renormalizations of excited e-h pairs and virtual e-h pairs in the condensate. Most importantly, the anharmonic exciton-photon interaction yields now an additional Stark contribution Π^F to both the shift and bleaching of the exciton resonances. This additional renormalization results from the composite nature of excitons, in much the same way as the exciton-exciton interaction.

If the pump detuning from the lowest $n=1s$ transition is not too large, most of the virtual e-h pairs will occupy this state, leading to a Stark shift of the $1s$ exciton, as measured by the test beam (only),[10]

$$\Pi_{1s\,1s}^F \simeq \frac{2|\mu E_p|^2}{E_{1s}^0 - \omega_p} \sum_{\mathbf{k}} |\phi_{1s\mathbf{k}}|^2 \phi_{1s,\mathbf{k}}^* \phi_{1s,\mathbf{r}=0} . \tag{67}$$

Under the same condition,[40]

$$n_{\mathbf{k}} = |\psi_{\mathbf{k}}|^2 \simeq \frac{N}{2}|\phi_{1s\mathbf{k}}|^2 . \tag{68}$$

Introducing a $1s$ exciton saturation density N_S^{PSF} due to PSF only,[40]

$$\chi_p^{\mathrm{PSF}}=\frac{2|\mu|^2}{E_{1s}^0-\omega_p}\sum_{\mathbf{k},\mathbf{k}'}\phi_{1s,\mathbf{k}}^*(1-2|\psi_{\mathbf{k}'}|^2)\phi_{1s,\mathbf{k}'}$$

$$=\frac{2|\mu|^2|\phi_{1s\mathbf{r}=0}|^2}{E_{1s}^0-\omega_p}\left[1-\frac{N}{N_S^{\mathrm{PSF}}}\right], \tag{69a}$$

where

$$N_S^{\mathrm{PSF}-1}=\sum_{\mathbf{k}}|\phi_{1s,\mathbf{k}}|^2\phi_{1s,\mathbf{k}}/\phi_{1s,\mathbf{r}=0}, \tag{69b}$$

Eq. (67) can be rewritten in the more intuitive form[3,10]

$$\Pi_{1s1s}^F\simeq\frac{2|\mu E_p|^2}{E_{1s}^0-\omega_p}\frac{|\phi_{1s,\mathbf{r}=0}|^2}{N_S^{\mathrm{PSF}}}. \tag{70}$$

This result should be contrasted with the Stark shift of the band gap in the case of noninteracting e and h, Eq. (56), or that of two-level atoms. The first factor in Eq. (70) expresses the Stark shift of the atomic s and p states that form the conduction and valence bands. The second factor describes the renormalization of this atomic shift due to excitonic effects. Its numerator reflects the fact that an exciton is built up from a linear combination of Bloch states that originate themselves from the atomic states. It expresses the enhancement of the oscillator strength due to the correlation in the excitonic state. The same factor appears in Elliott's formula for excitonic linear absorption. The denominator contains the saturation density N_S^{PSF}, above which the concept of excitons becomes invalid. Note that the magnitude of this second factor is rather independent of dimension; $|\phi_{1s,\mathbf{r}=0}|^2/N_S^{\mathrm{PSF}}=\frac{7}{2}$ and $\frac{16}{7}$ in three and two dimensions, respectively. If we identify Eqs. (69a) and (70) with the susceptibility and Stark shift of two-level atoms, we find that one exciton behaves like N_S^{PSF} independent two-level systems (if exciton-exciton interactions are neglected), i.e., N_S^{PSF} states are required to form an exciton. In the case of Frenkel excitons, we recover atomic behavior.

The exciton-exciton interaction can produce an additional blue shift, in much the same way as in the case of real excitons. While this shift is negligible in bulk semiconductors,[41] it becomes significant in narrow quantum wells which are close to ideal 2D behavior.[42,43] The physical origin of this behavior has been discussed elsewhere.[40,44]

Equation (70) explains already the recent experimental results obtained on 100 Å $GaAs/Al_xGa_{1-x}As$ quantum wells quantitatively, without any adjustable parameters. For a pump intensity $I_p\simeq 8\ \mathrm{MW\,cm^{-2}}$ and a pump detuning $E_{1s}^0-\omega_p\simeq 30$ meV, the magnitude of the experimental shift is 0.2 meV for the heavy-hole (hh) exciton peak and 0.05 for the light-hole (lh) exciton peak.[3] The theoretical hh exciton shift is 0.15 meV and the ratio of the hh exciton and lh exciton shifts 4, in excellent agreement with the experimental data.

Besides the Stark shift, there is of course a corresponding bleaching and, for higher pump intensities, there will also be optical gain, just like in the case of noninteracting e and h discussed above. This gain is again at least quadratic in the pump intensity and suitably interpreted in terms of the depletion of the condensate or simultaneous emission of a test photon and a collective excitation. From the diagonal part of Eq. (59) we obtain for the spectrum of collective excitations[10]

$$\omega_n=[(E_n^{\mathrm{HF}}-\omega_p)^2-|\Sigma_{nn}^B|^2]^{1/2}, \tag{71}$$

which is identical to the Bogolubov-Beliaev expression for the excitation spectrum of weakly nonideal Bose gas (but it has a gap).[20,21] Again, ω_p can be thought of as the quasichemical potential. Optical absorption into renormalized exciton states occurs for $\omega_t=\omega_p+\omega_n$ and optical gain for $\omega_t=\omega_p-\omega_n=2\omega_p-(\omega_p+\omega_n)$, i.e., at a frequency symmetric about ω_p.

For not too large pump detunings from the lowest exciton transition, and if we neglect the contribution to Eq. (71) of exciton-exciton interactions, i.e., if we keep only the self-energies $\Pi^{F,B}$ due to anharmonic exciton-photon interactions, Eq. (71) reduces to

$$\omega_{1s}^{\mathrm{PSF}}=[(E_{1s}^0+\Pi_{1s1s}^F-\omega_p)^2-|\Pi_{1s1s}^B|^2]^{1/2}. \tag{72}$$

Using $|\Pi_{1s1s}^B|=\Pi_{1s1s}^F$ and substituting Eq. (70), we find

$$\omega_{1s}^{\mathrm{PSF}}=[(E_{1s}^0-\omega_p)^2+4|\mu E_p|^2|\phi_{1s,\mathbf{r}=0}|^2/N_S^{\mathrm{PSF}}]^{1/2}, \tag{73}$$

which is exactly the expression for the Rabi frequency of a two-level system off resonance [see also Eq. (55)],[1] with suitable exciton modifications. For small pump intensities, this yields the linear Stark shift Eq. (70), while for large pump intensities saturation sets in. In this context, it should be noted, however, that for very high intensities it does no longer make sense to expand about unperturbed exciton states, so that Eq. (73) *cannot* be expected to describe the saturation of the Stark shift correctly.

Using the same formalism, one can also calculate the longitudinal dielectric function of virtually excited excitons, which describes the screening of external charges or fields. A test charge, $V_{\mathrm{ext}}(\mathbf{q},t)$, gives rise to a perturbation

$$\delta\hat{\varepsilon}_{\mathrm{ext}}(\mathbf{q},T)=V_{\mathrm{ext}}(\mathbf{q},T), \tag{74}$$

which combines with the local field due to induced charge density flucuations to give an effective potential

$$V_{\mathrm{eff}}(\mathbf{q},T)=V_{\mathrm{ext}}(\mathbf{q},T)+V_{\mathrm{ind}}(\mathbf{q},T), \tag{75}$$

where

$$V_{\mathrm{ind}}(\mathbf{q},T)=2V_q\sum_{\mathbf{k}}\mathrm{tr}\delta\hat{n}(\mathbf{k},\mathbf{q},T). \tag{76}$$

For small pump intensities and in the long-wavelength limit $q\to 0$, we obtain from Eq. (46), neglecting $n_{\mathbf{k}}$ in the denominator,

$$\lim_{q\to 0} V_{\rm ind}(\mathbf{q},T)\to 2V_q \sum_{\mathbf{k}} \left[\left[\mathbf{q}\cdot\frac{\partial\psi_{\mathbf{k}}}{\partial\mathbf{k}} \right] \lim_{q\to 0}\delta n_{21}(\mathbf{k},\mathbf{q},T) - \left[\mathbf{q}\cdot\frac{\partial\psi^*_{\mathbf{k}}}{\partial\mathbf{k}} \right] \lim_{q\to 0}\delta n_{12}(\mathbf{k},\mathbf{q},T) \right] , \tag{77}$$

and from Eq. (43)

$$\sum_{\mathbf{k}'} \left[\left[i\frac{\partial}{\partial T}+\omega_p \right]\delta_{\mathbf{k},\mathbf{k}'} - H^0_{\mathbf{k},\mathbf{k}'} \right] \lim_{q\to 0}\delta n_{12}(\mathbf{k}',\mathbf{q},T) \to - \left[\mathbf{q}\cdot\frac{\partial\psi_{\mathbf{k}}}{\partial\mathbf{k}} \right] \lim_{q\to 0} V_{\rm eff}(\mathbf{q},T) , \tag{78a}$$

$$\sum_{\mathbf{k}'} \left[\left[i\frac{\partial}{\partial T}-\omega_p \right]\delta_{\mathbf{k},\mathbf{k}'} + H^0_{\mathbf{k},\mathbf{k}'} \right] \lim_{q\to 0}\delta n_{21}(\mathbf{k}',\mathbf{q},T) \to - \left[\mathbf{q}\cdot\frac{\partial\psi^*_{\mathbf{k}}}{\partial\mathbf{k}} \right] \lim_{q\to 0} V_{\rm eff}(\mathbf{q},T) . \tag{78b}$$

Expanding $\delta n_{12}(\delta n_{21})$ in terms of unperturbed exciton wave functions $\phi_{n\mathbf{k}}(\phi^*_{n\mathbf{k}})$, Eq. (29), and Fourier transforming Eqs. (75)–(78), we finally obtain for the longitudinal dielectric function

$$\begin{aligned}\lim_{q\to 0}\frac{\epsilon(\mathbf{q},\omega)}{\epsilon_0} &= \lim_{q\to 0}\frac{V_{\rm ext}(\mathbf{q},\omega)}{V_{\rm eff}(\mathbf{q},\omega)} \\ &\to 1-2V_q \sum_{n,\mathbf{k},\mathbf{k}'} \left[\frac{\left[\mathbf{q}\cdot\frac{\partial\psi^*_{\mathbf{k}}}{\partial\mathbf{k}}\phi_{n\mathbf{k}} \right]\left[\mathbf{q}\cdot\frac{\partial\psi_{\mathbf{k}'}}{\partial\mathbf{k}'}\phi^*_{n\mathbf{k}'} \right]}{\omega+i0-E^0_n+\omega_p} - \frac{\left[\mathbf{q}\cdot\frac{\partial\psi_{\mathbf{k}}}{\partial\mathbf{k}}\phi^*_{n\mathbf{k}} \right]\left[\mathbf{q}\cdot\frac{\partial\psi^*_{\mathbf{k}'}}{\partial\mathbf{k}'}\phi_{n\mathbf{k}'} \right]}{\omega+i0+E^0_n-\omega_p} \right] \\ &= 1-4V_q \sum_n \frac{|\langle n|\mathbf{q}\cdot\mathbf{r}|\psi\rangle|^2(E^0_n-\omega_p)}{(\omega+i0)^2-(E^0_n-\omega_p)^2} ,\end{aligned} \tag{79}$$

where ϵ_0 is the background dielectric constant. Equation (79) shows once more that the long-wavelength excitation spectrum in the low-intensity limit is simply $\omega_n = E^0_n - \omega_p$, i.e., the energy required to create real excitons.

The consistency of our approximations is easily proven by noting that the dielectric function Eq. (79) fulfills the f-sum rule exactly. For large frequencies ω, and using Eqs. (36) and (37), we find

$$\begin{aligned}\lim_{\substack{q\to 0\\ \omega\to\infty}}\frac{\epsilon(\mathbf{q},\omega)}{\epsilon_0} &= 1-\frac{4V_q}{\omega^2}\sum_n |\langle n|\mathbf{q}\cdot\mathbf{r}|\psi\rangle|^2(E^0_n-\omega_p) \\ &= 1-\frac{4V_q}{\omega^2}\frac{q^2}{2m}\langle\psi|\psi\rangle \\ &= 1-\frac{NV_q q^2}{m\omega^2} ,\end{aligned} \tag{80}$$

which yields the exact plasma frequency both in 2D and 3D [$V^{\rm 2D}_q=(2\pi e^2)/(\epsilon_0 q)$, $V^{\rm 3D}_q=(4\pi e^2)/(\epsilon_0 q^2)$].

For almost resonant excitation of $1s$ excitons, $\omega_p \simeq E^0_{1s}$, Eq. (79) reduces to[7]

$$\lim_{q\to 0}\frac{\epsilon(\mathbf{q},\omega)}{\epsilon_0} \simeq 1-2NV_q \sum_n \frac{|\langle n|\mathbf{q}\cdot\mathbf{r}|1s\rangle|^2(E^0_n-E^0_{1s})}{(\omega+i0)^2-(E^0_n-E^0_{1s})^2} , \tag{81}$$

which is nothing but the dynamical polarizability of hydrogen atoms. Together with our other results, Eq. (81) clearly demonstrates that *all* collective properties of condensed excitons can easily be derived from the gauge invariant formulation of BCS-type pairing theory for the underlying fermions. Previous approaches to screening in condensed exciton systems[26,28] are not correct, because local-field corrections are neglected, so that the Ward identities are violated. Much the same can be said for a recent discussion of the saturated state of a semiconductor.[34]

If we replace ω_p by the quasichemical potential, i.e., E^H_{1s} in the low-density limit, and $n_{\mathbf{k}}$ by the quasiequilibrium distribution function Eq. (20) (with renormalized parameters), and let the symmetry breaking pump field go to zero, our formalism describes of course real Bose condensed excitons as well. The excitation spectrum is now gapless and, if one allows for a finite center momentum $\mathbf{q}$ in Eq. (48), one readily finds that $\lim_{q\to 0}\omega_{\mathbf{q}}=cq$, with a density-dependent sound velocity, which interpolates smoothly between the Anderson-Bogoliubov mode[17,18] at high densities and the Bogoliubov mode[20] at low densities. Both the ground state of Bose condensed excitons *and* their collective properties can thus be described in the framework of one and the same model, which reduces to the Bogoliubov-Beliaev theory of a weakly nonideal Bose gas in one limit and ordinary weak coupling BCS theory in the other. Much the same can be said for superconductors. What is still missing is a calculation of the critical temperature for arbitrary coupling,[29] which will be reported elsewhere.

V. CONCLUSIONS

We have presented a nonequilibrium theory of the coherent nonlinear optical response of semiconductors strongly driven by an intense monochromatic laser field and in the collisionless regime. This theory correctly accounts, for the first time, for the strong mutual interac-

tions of the extended electronic excitations in semiconductors. The interband interaction, i.e., the e-h attraction, is found to renormalize the Rabi frequency, while the intraband interaction, i.e., the e-e and h-h Coulomb repulsion, produces the usual renormalization of the e and h energy bands. The theory extrapolates well from real-space pairing (excitons) in the case of moderate pump intensities to momentum-space pairing (e-h Cooper pairs) in the case of very large pump intensities. Virtual excitons behave like a driven condensate of interacting weakly nonideal bosons. The excitons have an anharmonic interaction among themselves that can be interpreted as arising from mutual polarization and hard-core repulsion. The interaction of excitons with photons is anharmonic as well. This anharmonicity is directly determined by the filling of the phase space and has its roots in the underlying Fermi statistics obeyed by the components of composite bosons.

We have also determined the linear response of the renormalized system to weak perturbations produced by a test photon field or a test charge. In the dilute limit, we obtain analytical expressions which have simple and meaningful physical interpretations and which compare very well with the available experimental results.

Throughout the paper, we have stressed the fundamental similarities between the correct description of photoexcited semiconductors and the theories developed to describe coherent phenomena in other extended and interacting electronic systems, such as superfluids and superconductors. The present work demonstrates once more the close relationship between quantum optics and low-temperature physics. The fundamental concepts developed in condensed matter physics explain of course the behavior of localized and noninteracting atomic systems as well, as a limiting case. On the other hand, many interesting effects which have been observed and investigated first in atomic physics are just starting to be studied in the condensed state. Photoexcited semiconductors represent a key system in which the two fields merge completely. However, some of the ideas are now being applied to quantum fluids as well, as demonstrated by the recent discussion of self-induced transparency of sound waves in ^{3}He.[45]

ACKNOWLEDGMENTS

Part of this work was performed while one of us (H.H.) was visiting AT&T Bell Laboratories. He wishes to thank the technical staff for their hospitality.

[1]For a review, see S. Feneuille, Rep. Prog. Phys. **40**, 1257 (1977).
[2]A. Mysyrowicz, D. Hulin, A. Antonetti, A. Migus, W. T. Masselink, and H. Morkoc, Phys. Rev. Lett. **56**, 2748 (1986).
[3]A. Von Lehmen, J. E. Zucker, J. P. Heritage, and D. S. Chemla, Opt. Lett. **11**, 609 (1986).
[4]K. Tai, J. Hegarty, and W. T. Tsang. Appl. Phys. Lett. **51**, 152 (1987).
[5]J. L. Oudar, D. Hulin, A. Migus, A. Antonetti, and F. Alexandre, Phys. Rev. Lett. **55**, 2074 (1985).
[6]W. H. Knox, C. Hirlimann, D. A. B. Miller, J. Shah, D. S. Chemla, and C. V. Shank, Phys. Rev. Lett. **56**, 1191 (1986).
[7]For reviews, see H. Haug and S. Schmitt-Rink, Prog. Quant. Electron. **9**, 3 (1984), and J. Opt. Soc. Am. B **2**, 1135 (1985), and the recent collection of articles in *Optical Nonlinearities and Instabilities in Semiconductors,* edited by H. Haug (Academic, New York, 1987).
[8]L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1515 (1964) [Sov. Phys.—JETP **20**, 1018 (1965)].
[9]L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (Benjamin, New York, 1962).
[10]S. Schmitt-Rink and D. S. Chemla, Phys. Rev. Lett. **57**, 2752 (1986).
[11]A. L. Ivanov and L. V. Keldysh, Zh. Eksp. Teor. Fiz. **84**, 404 (1982) [Sov. Phys.—JETP **57**, 234 (1983)].
[12]H. Haug, J. Lumin. **30**, 171 (1985), and in Ref. 7.
[13]W. Schaefer and J. Treusch, Z. Phys. B **63**, 407 (1986).
[14]G. D. Mahan, Phys. Rep. **145**, 251 (1987).
[15]J. Rammer and H. Smith, Rev. Mod. Phys. **58**, 323 (1986).
[16]J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).
[17]P. W. Anderson, Phys. Rev. **112**, 1900 (1958).
[18]N. N. Bogolubov, V. V. Tolmachev, and D. V. Shirkov, *A New Method in the Theory of Superconductivity* (Academy of Sciences of the U.S.S.R., Moscow, 1958).
[19]O. Betbeder-Matibet and P. Nozières, Ann. Phys. **51**, 392 (1969).
[20]N. N. Bogolubov, J. Phys. USSR **11**, 23 (1947).
[21]S. T. Beliaev, Zh. Eksp. Teor. Fiz. **34**, 417 (1958); **34**, 433 (1958) [Sov. Phys.—JETP **7**, 289 (1958); **7**, 299 (1958)].
[22]E. P. Gross, Nuovo Cimento **20**, 454 (1961).
[23]L. P. Pitaevskii, Zh. Eksp. Teor. Fiz. **36**, 1168 (1959); **40**, 646 (1961) [Sov. Phys.—JETP **9**, 830 (1959); **13**, 451 (1961)].
[24]L. V. Keldysh and Yu. V. Kopaev. Fiz. Tverd. Tela **6**, 2791 (1964) [Sov. Phys.-Solid State **6**, 2219 (1965)].
[25]L. V. Keldysh and A. N. Kozlov, Zh. Eksp. Teor. Fiz. **54**, 978 (1968) [Sov. Phys.—JETP **27**, 521 (1968)].
[26]R. Zimmermann, Phys. Status Solidi B **76**, 191 (1976).
[27]A. P. Silin, Fiz. Tverd. Tela **19**, 134 (1977) [Sov. Phys.-Solid State **19**, 77 (1977)].
[28]C. Comte and P. Nozières, J. Phys. (Paris) **43**, 1069 (1982); **43**, 1083 (1982).
[29]P. Nozières and S. Schmitt-Rink, J. Low. Temp. Phys. **59**, 195 (1985).
[30]V. M. Galitskii, S. P. Goreslavskii, and V. F. Elesin, Zh. Eksp. Teor. Fiz. **57**, 207 (1969) [Sov. Phys.—JETP **30**, 117 (1970)].
[31]V. F. Elesin, Zh. Eksp. Teor. Fiz. **59**, 602 (1970) [Sov. Phys.—JETP **32**, 328 (1971)].
[32]A. S. Aleksandrov and V. F. Elesin, Fiz. Tekh. Poluprov. **5**, 615 (1971) [Sov. Phys.-Semicond. **5**, 542 (1971)].
[33]Th. Harbich and G. Mahler, Phys. Status Solidi B **104**, 185 (1981).
[34]C. Comte and G. Mahler, Phys. Rev. B **34**, 7164 (1986).
[35]R. J. Elliott, Phys. Rev. **108**, 1384 (1957).
[36]S. Schmitt-Rink (unpublished).
[37]P. Doerre, H. Haug, and D. B. Tran Thoai, J. Low. Temp. Phys. **35**, 465 (1979).
[38]P. Nozières and D. Saint James, J. Phys. (Paris) **43**, 1133 (1982).
[39]For a review, see F. Henneberger, Phys. Status Solidi B **137**, 371 (1986).

[40]S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Phys. Rev. B **32**, 6601 (1985).
[41]L. Schultheis, J. Kuhl, A. Honold, and C. W. Tu, Phys. Rev. Lett. **57**, 1635 (1986).
[42]N. Peyghambarian, H. M. Gibbs, J. L. Jewell, A. Antonetti, A. Migus, D. Hulin, and A. Mysyrowicz, Phys. Rev. Lett. **53**, 2433 (1984).
[43]D. Hulin, A. Mysyrowicz, A. Antonetti, A. Migus, W. T. Masselink, H. Morkoc, H. M. Gibbs, and N. Peyghambarian, Phys. Rev. B **33**, 4389 (1986).
[44]S. Schmitt-Rink, *Theory of Transient Excitonic Nonlinearities in Quantum Wells,* Proceedings of the National Science Foundation Workshop on Optical Nonlinearities, Fast Phenomena and Signal Processing (Optical Sciences Center, Tucson, 1986) p. 242.
[45]T. Kopp and P. Woelfle, Phys. Rev. Lett. (to be published).