Quasibosonic exciton dynamics near the semiconductor band edge

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We develop a heirarchy of equations that describes the electrodynamics of the semiconductor band edge. An exciton basis that incorporates intrapair Coulomb interaction is used. The excitons are treated as quasibosons that satisfy commutation relations intermediate between bosonic and fermionic. Their nonlinear dynamics are a consequence of phase-space filling and an effective exciton-exciton interaction that depends only on their center-of-mass wave vectors and internal quantum numbers. The resulting expansion in powers of the optical field is analogous to Axt and Stahl's dynamically controlled truncation, but takes a simpler form. If damping is omitted or if all interband and intraband dephasing time constants are the same, we find a set of relationships among the interband and intraband correlation functions which in the free pair basis give the semiconductor Bloch equations. [S0163-1829(98)06107-4]

I. INTRODUCTION

Semiconductor Bloch equations (SBE's) (Ref. 1) provide a successful and widely used description of the electrodynamics of the semiconductor band edge. The random-phase approximation is used to reduce the expectation value of the four-operator Coulomb terms to products of two operator terms that describe interband polarization and the numbers of free electrons and holes. Recently a number of theoretical and experimental papers discussed the limitations of these equations. It was demonstrated that the SBE's are based on an ill-controlled Hartree-Fock approximation, and that the band edge should be described by an infinite heirarchy of electron correlation functions.[2,3] This heirarchy can be systematically terminated using a dynamically controlled truncation (DCT) scheme that is an expansion in powers of the optical field.

Even in second order, problems arise with the SBE's because the factorization of higher-order correlations into polarization and number parts gets some of the decay time constants wrong. The Coulomb coupling of the electron and hole within a pair in the SBE's decays faster than the intraband polarization,[3] since factorization implies that intraband processes decay twice as fast as interband processes. The SBE's thus predict THz emission with frequencies that are characteristic of free electrons and holes. Experiment[4] has verified the prediction[3,5] that in fact THz emission is dominated by excitonic processes.

In contrast to Ref. 2, we work in a basis of Wannier excitons whose centers of mass are free to move throughout the crystal. Formation of an exciton can be thought of as destruction of an electron in the valence band, leaving a hole, followed by creation of an electron in the conduction band. In a second-quantized formalism, the process is described mathematically by an exciton creation operator which is a sum of products of fermionic electron and hole creation operators. Excitons are bosons at low densities, and their creation and destruction operators, can be shown to satisfy bosonic commutation relations to a first approximation.[6] This is in contrast to the SBE which is primarily formulated in **k** space, where the free electrons and holes have fermionic state filling properties.

We develop a theory of the dynamics of the semiconductor band edge that it is based on exciton operators in an ordered pair space. We work in the product space[7] of fermions and quasibosons (qbosons) and transform the fermion commutation relations and Hamiltonian for electrons and holes to the quasibosonic space of the excitons. However, the dynamical equations that we derive utilize qboson commutation relations that allow for phase-space filling. The Hamiltonian that we obtain is essentially identical to that used by Hanamura and Haug to describe strictly bosonic excitons except that we include coupling to the optical field. Equations of motion for the exciton operators are derived in the Heisenberg picture.

Expectation values that describe interband, intraband, and interband-intraband polarization and numbers are then defined using products of the exciton creation and annihilation operators. The expansion in powers of the number of exciton operators is found to be equivalent to an expansion in powers of the optical field, $\mathcal{E}$, and analogous to the DCT scheme of Axt and co-workers.[2,3] The use here of ordered electron-hole pairs in a qboson space simplifies the expansion. To order $\mathcal{E}^4$, the expectation values of the coherent interband response and the number operators without damping give the SBE.

The plan of the paper is as follows. In Sec. II we derive the exciton commutation relations using Usui's transform.[7] In Sec. III we derive, in a like manner, the Hamiltonian for a system of excitons interacting with each other and coupled to the field. The Heisenberg dynamical equations for the exciton operators are also obtained. In Sec. IV we introduce dephasing in the interaction picture and derive equations for the expectation values of the interband, intraband, and interband-intraband operators. Finally we conclude with a discussion of DCT and SBE's and the physical significance of these results.

II. COMMUTATION RELATIONS

In this section, commutation relations for creation and annihilation operators acting in the space of spinless

electron-hole pairs will be derived using Usui's transformation[7] from fermion to boson space. It will be assumed that electrons and holes exist only as pairs, which is the case if they are created optically. A linear transformation to a different set of operators will be made, and their commutation relations will be derived. The term exciton is used here to denote both bound and unbound electron-hole pairs in the new basis.

In a two-band model with one conduction band and one valence band, the operator $\alpha_{\mathbf{k}}$ annihilates an electron with wave vector $\mathbf{k}$, while $\beta_{\mathbf{k}}$ similarly destroys a hole. The operator that annihilates a pair with center-of-mass wave vector $\mathbf{K}$ and electron-hole relative wave vector $\mathbf{k}$ as defined in Ref. 1 is then

$$b_{\mathbf{k},\mathbf{K}}=\beta_{-\mathbf{k}+\alpha_h\mathbf{K}}\alpha_{\mathbf{k}+\alpha_e\mathbf{K}}. \tag{1}$$

Here m_e, m_h, and M are the electron, hole, and total masses, respectively, $\alpha_e\equiv m_e/M$ and $\alpha_h\equiv m_h/M$. Where it is unambiguous, the collective indices $k_i=\{\mathbf{k}_i,\mathbf{K}_i\}$ will be used to describe the state of the ith pair. The operators $b_{k_i}^\dagger$ (b_{k_i}) thus create (annihilate) an electron and hole with center-of-mass wave vector $\mathbf{K}_i$ and relative wave vector $\mathbf{k}_i$ or, equivalently, electron wave vector $\mathbf{k}_{\mathrm{ei}}$ and hole wave vector $\mathbf{k}_{\mathrm{hi}}$. Fermionic operator algebra then gives the commutation relations for the pair operators

$$[b_{k_1},b_{k_2}]=[b_{k_1}^\dagger,b_{k_2}^\dagger]=0 \tag{2}$$

and

$$[b_{k_1},b_{k_2}^\dagger]=\delta_{k_1,k_2}-\delta_{\mathbf{k}_{h1},\mathbf{k}_{h2}}\alpha_{\mathbf{k}e_2}^\dagger\alpha_{\mathbf{k}e1}-\delta_{\mathbf{k}e_1,\mathbf{k}e_2}\beta_{\mathbf{k}_{h2}}^\dagger\beta_{\mathbf{k}_{h1}}. \tag{3}$$

From Eq. (2), it follows that states created from the vacuum using $b_{k_i}^\dagger$ are symmetric under an exchange of pairs. This is a bosonic property that is to be expected for exchange of two fermions. However, Eq. (3) implies that state filling follows fermionic rules for these noninteracting electron-hole pairs.

A commutator of annihilation and creation operators such as $[b_{k_1},b_{k_2}^\dagger]$ essentially compares the creation of a particle with quantum numbers k_2 before and after annihilation of a particle with quantum numbers k_1. If the particle's phase-space filling properties are bosonic, then the presence of k_1 does not matter, and the commutator equals the Kronecker delta. For fermions, creation of a second particle is completely blocked if the state is already full. The form of Eq. (3) ensures that a pair k_2 cannot be created before annihilation of k_1, unless both the electron and hole states are empty.

It has been assumed that the operators act in a space that contains an equal number of electrons and holes. A fermionic state consisting of n electrons and n holes does not make any allowance for pairing. For example, the state containing electron-hole pairs with wave vectors $(\mathbf{k}_1,-\mathbf{k}_1;\mathbf{k}_2,-\mathbf{k}_2)$ is equivalent to the fermion state $(\mathbf{k}_2,-\mathbf{k}_1;\mathbf{k}_1,-\mathbf{k}_2)$, since they both contain the same two free electrons and free holes. However, they can be viewed as different exciton states, and only the former consists of two pairs each with zero center-of-mass motion. It is important to maintain this distinction in the discussion of hydrogenlike excitons, since all the pairs that contribute to a given exciton have a common center-of-mass motion. There is a one-to-$n!$ correspondence between fermion space, and the corresponding boson space where permutations of the electrons for fixed holes produce $n!$ nonequivalent states.

Since the excitons to be considered are not ideal bosons, the space of paired electrons and holes will here be called pair space or qboson space. In pair space, states are symmetric under exchange of pairs, but not necessarily antisymmetric under an exchange of individual electrons and holes. Fermion exchange is included in the energy of individual excitons and in the exciton-exciton interaction energies. Any two states containing different numbers of electrons and holes or a different $\{k_i\}$ are orthogonal. However, the ordering of the $k_i's$ does not matter, since the state is symmetric under an exchange of pairs.

To perform the transformation from the fermionic electron and hole operators to pair operators, Usui's transformation[7] (also see Ref. 6) will be used. The operator

$$U=P_F\,\exp\left(\sum_{\mathbf{k}_e,\mathbf{k}_h}B_{\mathbf{k}_e,\mathbf{k}_h}^\dagger b_{\mathbf{k}_e,\mathbf{k}_h}\right)P_B \tag{4}$$

acts in the product space of fermions and qboson pairs where P_F and P_B are projection operators onto the fermion and qboson vacuums, $|0\rangle_F$ and $|0\rangle_B$, respectively. The state $|0\rangle\equiv|0\rangle_F|0\rangle_B$, containing no electrons, holes, or pairs, will be called the vacuum here. The operator b that annihilates an electron and a hole in the fermion subspace of the product space conveys no information about pairing, while $B^\dagger$ creates a pair in the qboson subspace. When U acts on a fermion state, it converts it to the antisymmetrized linear combination of pairs.[7]

To obtain a one-to-one correspondence between the fermion states and a certain subspace of the boson states, Usui introduced an ordering operator, O, whose eigenvalue is 1 if the (electron-hole) pairing is in compliance with an ordering prescription, and 0 otherwise. Then OU transforms a fermion state to this boson subspace, where O is a projection operator satisfying $O^2=O$, giving $U^\dagger OU=1$ which implies that OU is unitary. A possible choice would be to pair an electron with wave vector $\mathbf{k}$ with a hole with wave vector $-\mathbf{k}$; however, the particular form of the pairing prescription does not matter in what follows, since pairing is not changed by the qboson operators. The fermion operator A_F will be transformed to a qboson operator A_B, using

$$A_B=OUA_FU^\dagger O. \tag{5}$$

Equation (5) can be used to convert the fermion operators in Eqs. (2) and (3) to qboson operators. Since zero transforms to zero, the symmetry relation (2) becomes

$$[B_{k_1},B_{k_2}]=[B_{k_1}^\dagger,B_{k_2}^\dagger]=0, \tag{6}$$

while the commutator of an annihilation operator with a creation operator is

$$[B_{k_1},B_{k_2}^\dagger]\equiv\delta_{k_1,k_2}-(\delta_{\mathbf{k}_1,\mathbf{k}_2+\alpha_h(\mathbf{K}_1-\mathbf{K}_2)}+\delta_{\mathbf{k}_1,\mathbf{k}_2-\alpha_e(\mathbf{K}_1-\mathbf{K}_2)})B_{k_2}^\dagger B_{k_1}. \tag{7}$$

If pair k_1 exists then it blocks the creation of k_2 provided they have a common electron or hole wave vector. Details of this transformation are given in the Appendix.

A set of exciton operators can be introduced as a linear combination of these pair operators,

$$B^{\dagger}_{n,\mathbf{K}}=\sum_{\mathbf{k}} \psi_{n,\mathbf{k}}B^{\dagger}_{\mathbf{k},\mathbf{K}}, \tag{8}$$

where $\psi_{n,\mathbf{k}}$ is the k-space representation of our basis, and periodic boundary conditions have been used. This transformation affects the relative motion of the electron and hole, but does not change the center of mass motion of the pair. Multiplying both sides of Eq. (8) by $\psi_{n,\mathbf{k}'}$, summing over n, and using the completeness of the basis gives the inverse transformation

$$B^{\dagger}_{\mathbf{k},\mathbf{K}}=\sum_{n} \psi^{*}_{n,\mathbf{k}}B^{\dagger}_{n,\mathbf{K}}, \tag{9}$$

with adjoint expressions for the $B's$. We also define a Greek collective index $\nu_i=\{n_i,\mathbf{K}_i\}$ that includes the quantum numbers of an exciton in our basis, and allows us to write the exciton creation operator as $B^{\dagger}_{\nu}$.

Using expansion (8) of the exciton operators in terms of plane-wave pair operators, Eq. (6) leads to

$$[B^{\dagger}_{\nu_1},B^{\dagger}_{\nu_2}]=[B_{\nu_1},B_{\nu_2}]=0. \tag{10}$$

The commutators of exciton annihilation and creation operators [Eq. (8)] become, after substitution of Eq. (7) and inverse transformation (9),

$$[B_{\nu_1},B^{\dagger}_{\nu_2}]=\delta_{\nu_1,\nu_2}-2\sum_{m_1,m_2} \chi^{n_1,n_2,\mathbf{K}_1-\mathbf{K}_2}_{m_1,m_2}B^{\dagger}_{\mu_2}B_{\mu_1}, \tag{11}$$

with $\nu_i=\{n_i,\mathbf{K}_i\}$, $\mu_i=\{m_i,\mathbf{K}_i\}$ and

$$\chi^{n_1,n_2,\mathbf{Q}}_{m_1,m_2}\equiv \tfrac{1}{2}\sum_{\mathbf{k}} (\psi^{*}_{n_1,\mathbf{k}}\psi_{n_2,\mathbf{k}+\alpha_h\mathbf{Q}}\psi^{*}_{m_2,\mathbf{k}+\alpha_h\mathbf{Q}}\psi_{m_1,\mathbf{k}} + \psi^{*}_{n_1,\mathbf{k}}\psi_{n_2,\mathbf{k}-\alpha_e\mathbf{Q}}\psi^{*}_{m_2,\mathbf{k}-\alpha_e\mathbf{Q}}\psi_{m_1,\mathbf{k}}). \tag{12}$$

These χ parameters describe phase-space filling, and can be calculated for any particular exciton basis. The effect of center of mass motion is to shift the $\psi_{n,\mathbf{k}}$ functions in $\mathbf{k}$ space, spreading them out and making room for more pairs. The parameter $\chi^{n_1,n_1,0}_{n_1,n_1}$ is the average probability for the occurrence of any given pair. If $\chi=0$, the excitons are bosons.

Commutation relations (10) and (11), with definition (12), define the properties of the exciton operators and the space in which they act. States created using $\{B^{\dagger}_{\nu_i}\}$ are symmetric under exchange of excitons. In the special case $\psi_{n,\mathbf{k}}=\delta_{\mathbf{k}_n,\mathbf{k}}$, the commutation relations for the pair operators (6) and (7) are regained.

III. HAMILTONIAN AND DYNAMICS

In this section the fermion Hamiltonian will be written in terms of qboson operators as in Ref. 6 and Heisenberg equations for the creation operators will be derived. The Hamiltonian of a system of electrons and holes interacting with an electromagnetic field should include contributions from the noninteracting particles, coupling between the particles and the field, and particle-particle Coulomb interactions. The starting point is a basis of noninteracting electrons and holes,[1,6] and its electron and hole operators, can first be transformed to pair space. Some details are given at the end of the Appendix. In center-of-mass and relative coordinates, the pair-space dipolar Hamiltonian becomes

$$H=H_0+H_I+H_C, \tag{13}$$

with

$$H_0=\sum_{k_1} E^{0}_{k_1}B^{\dagger}_{k_1}B_{k_1}-\sum_{\mathbf{q},k_1} V_qB^{\dagger}_{\mathbf{k}_1+\mathbf{q},\mathbf{K}_1}B_{k_1}, \tag{14}$$

$$H_I=-\sum_{k_1} \mathcal{E}_{\mathbf{K}_1}(t)(\mu_{\mathrm{cv}}B_{\mathbf{k}_1,\mathbf{K}_1}+\mu^{*}_{\mathrm{cv}}B^{\dagger}_{\mathbf{k}_1,-\mathbf{K}_1}), \tag{15}$$

$$H_C=H_P(k_1,k_2)-H_P(k_{E1},k_{E2}) + \sum_{\mathbf{q},k_1,k_2} V_qB^{\dagger}_{k_{E1}}B^{\dagger}_{\mathbf{k}_{E2}+\mathbf{q},\mathbf{K}_{E2}}B_{k_2}B_{k_1}, \tag{16}$$

where

$$H_P(k'_1,k'_2)\equiv \tfrac{1}{2}\sum_{\mathbf{q}\neq 0,k_1,k_2} V_q(B^{\dagger}_{\mathbf{k}'_1+\alpha_h\mathbf{q},\mathbf{K}'_1+\mathbf{q}} - B^{\dagger}_{\mathbf{k}'_1-\alpha_e\mathbf{q},\mathbf{K}'_1+\mathbf{q}})(B^{\dagger}_{\mathbf{k}'_2-\alpha_h\mathbf{q},\mathbf{K}'_2-\mathbf{q}} - B^{\dagger}_{\mathbf{k}'_2+\alpha_e\mathbf{q},\mathbf{K}'_2-\mathbf{q}})B_{k_2}B_{k_1},$$

$\mu_{cv}=\langle c|e\mathbf{r}\cdot\mathbf{e}_{\mathbf{K}}|v\rangle$, $\mathcal{E}_{\mathbf{K}}(t)$ is the time-dependent electric field at wave vector $\mathbf{K}$, the Fourier transformed Coulomb potential is V_q, and $E^{0}_{k_1}$ is the energy of a noninteracting electron-hole pair. In the pair interaction Hamiltonian H_P the first terms in brackets describe the scattered electrons, while the second term describes the scattered holes.

Both direct and exchange interactions contribute to the Coulomb Hamiltonian H_C. After exchange of electrons the relative and center-of-mass wave vectors become $\mathbf{k}_{E1}=\mathbf{k}_1+\alpha_h\mathbf{k}_{21}$, $\mathbf{K}_{E1}=\mathbf{K}_1+\mathbf{k}_{21}$, $\mathbf{k}_{E2}=\mathbf{k}_2-\alpha_h\mathbf{k}_{21}$, and $\mathbf{K}_{E2}=\mathbf{K}_2-\mathbf{k}_{21}$, with

$$\mathbf{k}_{21}\equiv\alpha_e(\mathbf{K}_2-\mathbf{K}_1)+\mathbf{k}_2-\mathbf{k}_1. \tag{17}$$

It is not necessary to exchange both electrons and holes, since this introduces no new pairs. The electron-electron and hole-hole interactions each generate two terms, while the electron-hole interaction transforms into four terms that contribute to H_C and generate the second term of H_0. In the coefficients, ϵ is the dielectric constant in the medium, d is the dimension, L is the linear system size, and c is the speed of light in the medium. The terms in the Hamiltonian describe single pairs, pair-field coupling, and the direct and exchange Coulomb interactions. If there is no external field, the above Hamiltonian is equivalent to the Hamiltonian obtained by Hanamura and Haug for bosonic excitons.[6]

Transformation to an exciton basis that incorporates the electron-hole Coulomb interaction within a pair gives, using Eq. (9),

$$H_0=\sum_{\nu_1} E^0_{\nu_1}B^\dagger_{\nu_1}B_{\nu_1}, \tag{18}$$

$$H_I=-\sum_{\nu_1}\mathcal{E}_{\mathbf{K}_1}(t)(\mu_{n_1}B_{n_1,\mathbf{K}_1}+\mu^*_{n_1}B^\dagger_{n_1,-\mathbf{K}_1}), \tag{19}$$

$$H_C=\sum_{\nu_1,\nu_2,n_3,n_4,\mathbf{Q}} W^{n_1,n_2,\mathbf{K}_1-\mathbf{K}_2}_{n_3,n_4,\mathbf{Q}}B^\dagger_{n_4,\mathbf{K}_1+\mathbf{Q}}B^\dagger_{n_3,\mathbf{K}_2-\mathbf{Q}}B_{\nu_2}B_{\nu_1}, \tag{20}$$

where the dipole moment is

$$\mu_n\equiv\mu_{\mathrm{cv}}\psi_n(\mathbf{r}=0)L^{d/2}.$$

The exciton pair potential is

$$\begin{aligned}W^{n_1,n_2,\mathbf{K}_1-\mathbf{K}_2}_{n_3,n_4,\mathbf{Q}}=&\sum_{\mathbf{q}\neq 0,\mathbf{k}_1,\mathbf{k}_2} V_q[\,\delta_{\mathbf{q},\mathbf{Q}}F^{n_3,n_4}_{\mathbf{q}}(\mathbf{k}_1,\mathbf{k}_2)\\&-\delta_{\mathbf{q},\mathbf{Q}-\mathbf{k}_{21}}F^{n_3,n_4}_{\mathbf{q}}(\mathbf{k}_1+\alpha_h\mathbf{k}_{21},\mathbf{k}_2-\alpha_h\mathbf{k}_{21})\\&+\delta_{\mathbf{k}_{21},\mathbf{Q}}\psi^*_{n_4,\mathbf{k}_1+\alpha_h\mathbf{k}_{21}}\psi^*_{n_3,\mathbf{k}_2-\alpha_h\mathbf{k}_{21}+\mathbf{q}}]\\&\times\psi_{n_2,\mathbf{k}_2}\psi_{n_1,\mathbf{k}_1},\end{aligned} \tag{21}$$

with

$$\begin{aligned}F^{n_3,n_4}_{\mathbf{q}}(\mathbf{k}'_1,\mathbf{k}'_2)\equiv&\tfrac{1}{2}(\psi^*_{n_4,\mathbf{k}'_1+\alpha_h\mathbf{q}}-\psi^*_{n_4,\mathbf{k}'_1-\alpha_e\mathbf{q}})\\&\times(\psi^*_{n_3,\mathbf{k}'_2-\alpha_h\mathbf{q}}-\psi^*_{n_3,\mathbf{k}'_2+\alpha_e\mathbf{q}}).\end{aligned} \tag{22}$$

The exciton energy $E^0_{n,K}$ includes the band gap E_g, the (negative) exciton energy E^0_n, and the center-of-mass energy, so that

$$E^0_\nu=E_g+E^0_n+\hbar^2K^2/2M.$$

The Coulomb interaction energy W for two excitons includes exchange, but once the total interaction energy has been calculated the details of exchange can be ignored in the exciton basis. It is included in the Hamiltonian rather than in the states, whose pairs are selected according to the ordering prescription and are not exchanged. If their spins are the same, as modelled by the system of spinless particles discussed here, the excitons interact repulsively due to Pauli exclusion.

In the Heisenberg picture the dynamics of the operators is described by $i\hbar dA/dt=[A,H]$. For the exciton creation operators that satisfy Eq. (11), Hamiltonian (13) with Eqs. (18)–(20) gives

$$\begin{aligned}-i\hbar\frac{d}{dt}B^\dagger_\nu=&E^0_\nu B^\dagger_\nu-\sum_{\nu_1}\mu_n\mathcal{E}_{\mathbf{K}}(t)\\&\times\left(\delta_{\nu_1,\nu}-2\sum_{m_1,m}\chi^{n_1,n,\mathbf{K}_1-\mathbf{K}}_{m_1,m}B^\dagger_\mu B_{\mu_1}\right)\\&+\sum_{\nu_1,n_3,n_4,\mathbf{Q}}\widetilde{W}^{n_1,n,\mathbf{K}_1-\mathbf{K}}_{n_3,n_4,\mathbf{Q}}B^\dagger_{n_4,\mathbf{K}_1+\mathbf{Q}}B^\dagger_{n_3,\mathbf{K}-\mathbf{Q}}B_{\nu_1}\\&+X_\nu.\end{aligned} \tag{23}$$

The effective exciton-exciton Coulomb interaction including the phase-filling correction to the exciton energy is

$$\widetilde{W}^{n_1,n,\mathbf{P}}_{n_3,n_4,\mathbf{Q}}\equiv W^{n_1,n,\mathbf{P}}_{n_3,n_4,\mathbf{Q}}-2\delta_{\mathbf{Q},0}E^0_{\nu_1}\chi^{n_4,n,\mathbf{P}}_{n_3,n_1}, \tag{24}$$

and

$$\begin{aligned}X_\nu\equiv&\sum_{\nu_1,n_3,n_4,\mathbf{Q},\nu_2,m_2,m}W^{n_1,n_2,\mathbf{K}_1-\mathbf{K}}_{n_3,n_4,\mathbf{Q}}\chi^{n_2,n,\mathbf{K}_2-\mathbf{K}}_{m_2,m}B^\dagger_{n_4,\mathbf{K}_1+\mathbf{Q}}\\&\times B^\dagger_{n_3,\mathbf{K}_2-\mathbf{Q}}(B_{\nu_1}B^\dagger_{\mu_2}B_\mu+B^\dagger_{\mu_2}B_\mu B_{\nu_1})\end{aligned}$$

is a small phase-space filling correction to the Coulomb term. The first line of Eq. (23) includes the zero-order terms and an external driving field corrected for phase-space filling, while the second line incorporates all exciton-exciton interactions. The phase-space-filling χ factors are small for bound hydrogenlike excitons. Phase-space-filling effects are missed if the excitons are treated strictly as bosons ($\chi=0$), as for example in Ref. 6, but can be comparable in magnitude to the Coulomb terms.

Since phase-space filling has its origin in the fermionic properties of the free electrons and holes, its effects are sometimes best understood in **k** space. In Eqs. (24) and (23), the phase-space-filling correction to the exciton energy can be converted back to the original free pair basis using Eq. (9). This gives

$$\Sigma\mathbf{k}_1,n_1E^0_{\nu_1}\psi^*_{n_1,\mathbf{k}_1}\psi_{n,\mathbf{k}_1}B^\dagger_{n_1,\mathbf{K}_1}B^\dagger_{\mathbf{k}_1,\mathbf{K}}B_{\mathbf{k}_1,\mathbf{K}_1},$$

which can be written as

$$\Sigma_{\mathbf{k}_1,\mathbf{k}_2}H^0_{\mathbf{k}_1,\mathbf{k}_2}\psi_{n,\mathbf{k}_1}B^\dagger_{\mathbf{k}_1,\mathbf{K}_1}B^\dagger_{\mathbf{k}_1,\mathbf{K}}B_{\mathbf{k}_1,\mathbf{K}_1},$$

where $H^0_{\mathbf{k},\mathbf{k}'}$ is the **k**-space representation of H_0. For modes coupled to the field, **K** and $\mathbf{K}_1$ are approximately zero. Since $B^\dagger_{\mathbf{k},0}B^\dagger_{\mathbf{k},0}=0$, the nondiagonal terms in the exciton Hamiltonian contribute zero, leaving only the potential-energy contribution to H^0 which equals $V_{|\mathbf{k}_1-\mathbf{k}_2|}$.

IV. DENSITY MATRICES AND DEPHASING

In this section, equations of motion for the expectation values of the interband, intraband, and interband-intraband operators and the biexciton operator will be obtained to fourth order in the field. For comparison with experiment, it is necessary to introduce dephasing, usually through phenomenological decay constants. The dephasing of coherent polarization is described by decay of the off-diagonal terms in the density matrix. It is not possible to introduce decay directly into the operators, since the commutation relations would not be preserved. For another perspective, if the energy has an imaginary part describing decay, then the transformation from the Schrödinger to the Heisenberg picture is not unitary.

Here we will define H_{coh} as that part of the Hamiltonian that does not decouple modes from the optical field by changing their center-of-mass motion. The remainder of the Hamiltonian, called H_{int}, is responsible for dephasing and any other effects that involve change of center-of-mass momentum, where the total Hamiltonian is $H=H_{\mathrm{coh}}+H_{\mathrm{int}}$. Interactions with acoustic and optical phonons and defects can

also be included in $H_{\rm int}$. Then $U=e^{iH_{\rm coh}t/\hbar}$ transforms the operators to the interaction rather than the Heisenberg picture, and the state vector evolves according to $(\partial/\partial t)\,|\psi(t)\rangle = H_{\rm int}|\psi(t)\rangle$. The dynamical equations of the operators will be replaced by $i\hbar dA/dt=[A,H_{\rm coh}]$, so that the effective Coulomb term in Eq. (23) is reduced to the $\mathbf{Q}=0$ terms. If the difference in the initial center-of-mass wave vectors is also zero, the effective interaction (24) simplifies to

$$\widetilde{W}^{n_1,n,0}_{n_3,n_4,0}=2\sum_{\mathbf{k}_1,\mathbf{q}\neq 0} V_q(\psi^*_{n_4,\mathbf{k}_1+\mathbf{q}}\psi_{n,\mathbf{k}_1}\psi^*_{n_3,\mathbf{k}_1}\psi_{n_1,\mathbf{k}_1} - \psi^*_{n_4,\mathbf{k}_1}\psi^*_{n_3,\mathbf{k}_1+\mathbf{q}}\psi_{n,\mathbf{k}_1+\mathbf{q}}\psi_{n_1,\mathbf{k}_1}), \tag{25}$$

and the phase-space filling factor becomes

$$\chi^{n_1,n_2,0}_{m_1,m_2}=\sum_{\mathbf{k}}\psi^*_{n_1,\mathbf{k}}\psi_{n_2,\mathbf{k}}\psi^*_{m_2,\mathbf{k}}\psi_{m_1,\mathbf{k}}. \tag{26}$$

For zero change in center-of-mass motion, only the electron-electron and hole-hole interactions with exchange contribute to the exciton-exciton Coulomb interaction.

It is clear from the above arguments that, before introducing decay, it is necessary to take expectation values. The physical quantities of interest are the polarization and the numbers. The polarization is

$$\mathcal{P}=\int d^dr\langle\psi^\dagger(\mathbf{r})(-e\mathbf{r}\cdot\mathbf{e}_{\mathbf{K}})\psi(\mathbf{r})\rangle,$$

where the operator

$$\psi(\mathbf{r})=\sum_{b,\mathbf{k}} a_{b,\mathbf{k}}\psi_{b,\mathbf{k}}(\mathbf{r})$$

annihilates an electron in band b at $\mathbf{r}$. Using the transformations of $\alpha^\dagger_{\mathbf{k}}\alpha_{\mathbf{k}'}$ and $\beta^\dagger_{\mathbf{k}}\beta_{\mathbf{k}'}$ in the Appendix, this becomes, in the pair-space basis,

$$\mathcal{P}=\sum_{\mathbf{k}_e,\mathbf{k}_h}(\langle B^\dagger_{\mathbf{k}_e,\mathbf{k}_h}\rangle\mu_{\rm cv}+\langle B_{\mathbf{k}_e,\mathbf{k}_h}\rangle\mu^*_{\rm cv}) + \sum_{\mathbf{k}_e,\mathbf{k}'_e,\mathbf{k}_h}\langle B^\dagger_{\mathbf{k}_e,\mathbf{k}_h}B_{\mathbf{k}'_e,\mathbf{k}_h}\rangle\langle\mathbf{k}_e,\mathbf{k}_h|-e\mathbf{r}\cdot\mathbf{e}_{\mathbf{K}}|\mathbf{k}'_e,\mathbf{k}_h\rangle + \sum_{\mathbf{k}_e,\mathbf{k}'_h,\mathbf{k}_h}\langle B_{\mathbf{k}_e,\mathbf{k}_h}B^\dagger_{\mathbf{k}_e,\mathbf{k}'_h}\rangle\langle\mathbf{k}_e,\mathbf{k}_h|-e\mathbf{r}\cdot\mathbf{e}_{\mathbf{K}}|\mathbf{k}_e,\mathbf{k}'_h\rangle$$

or in the exciton basis, using Eq. (9),

$$\mathcal{P}=\sum_n(\mu_n\langle B^\dagger_{n,0}\rangle+\mu^*_n\langle B_{n,0}\rangle) + \sum_{\mathbf{k},\mathbf{k}',\mathbf{K},\mathbf{K}',n,n'}\psi_{n,\mathbf{k}}\psi^*_{n',\mathbf{k}'}(\delta_{\mathbf{k}',\mathbf{k}+\alpha_h(\mathbf{K}-\mathbf{K}')}\langle B^\dagger_{n,\mathbf{K}}B_{n',\mathbf{K}'}\rangle + \delta_{\mathbf{k}',\mathbf{k}-\alpha_e(\mathbf{K}-\mathbf{K}')}\langle B_{n,\mathbf{K}}B^\dagger_{n',\mathbf{K}'}\rangle)\langle\mathbf{k},\mathbf{K}|-e\mathbf{r}\cdot\mathbf{e}_{\mathbf{K}}|\mathbf{k}',\mathbf{K}'\rangle. \tag{27}$$

These expressions include interband and intraband contributions. The annihilation and creation operators B_ν and $B^\dagger_\nu$ will be referred to as interband operators, while $B^\dagger_\nu B_\mu$ and $B_\nu B^\dagger_\mu$ will be called intraband operators. If $\mu=\nu$, the former intraband product is a number operator. It is the expectation values of these operators that are of interest since, if they are known, the polarization can be found using Eq. (27).

Expectation values are taken with respect to $|\psi(t)\rangle$, whose evolution is determined by $H_{\rm int}$. From (23), one obtains

$$\left(i\frac{d}{dt}+\omega^0_\nu+\frac{i}{T_\nu}\right)\langle B^\dagger_\nu\rangle = \mathcal{R}_\nu-2\sum_{\nu_1,m_1,m}\chi^{n_1,n,\mathbf{K}_1-\mathbf{K}}_{m_1,m}\mathcal{R}_{\nu_1}\langle B^\dagger_\mu B_{\mu_1}\rangle - \sum_{\nu_1,n_3,n_4}\widetilde{W}^{n_1,n,\mathbf{K}_1-\mathbf{K}}_{n_3,n_4,0}\langle B^\dagger_{n_4,\mathbf{K}_1}B^\dagger_{n_3,\mathbf{K}}B_{\nu_1}\rangle+\mathcal{O}(\mathcal{R}^5), \tag{28}$$

where $\mathcal{R}_\nu\equiv\mu_n\mathcal{E}_{\mathbf{K}}(t)/\hbar$, $\omega^0_\nu\equiv E^0_\nu/\hbar$, and $T_\nu=T^{\rm inter}_2$ in Ref. 4. The intraband polarization is proportional to $\langle B^\dagger_\nu B_\mu\rangle$, and its equation of motion can also be derived from Eq. (23) by equating the time derivative of $B^\dagger_\nu B_\mu$ to $dB^\dagger_\nu/dtB_\mu + B^\dagger_\nu dB_\mu/dt$ and taking the expectation value of the resulting equation to obtain

$$i\frac{d}{dt}\langle B^\dagger_\nu B_\mu\rangle=-\left(\omega^0_{\nu\mu}+\frac{i}{T_{\nu\mu}}\right)\langle B^\dagger_\nu B_\mu\rangle+\mathcal{R}_\nu\langle B_\mu\rangle-\mathcal{R}^*_\mu\langle B^\dagger_\nu\rangle - 2\sum_{\nu_1,m_1,m_2}\{\chi^{n_1,n,\mathbf{K}_1-\mathbf{K}}_{m_1,m_2}\mathcal{R}_{\nu_1}\langle B^\dagger_{\mu_2}B_{\mu_1}B_\mu\rangle - \chi^{m_1,m_2,\mathbf{K}_1-\mathbf{K}'}_{n_1,n}\mathcal{R}^*_{\nu_1}\langle B^\dagger_\nu B^\dagger_{\mu_1}B_{\mu_2}\rangle\} - \sum_{\nu_1,n_1,n_3}\{\widetilde{W}^{n_1,n,\mathbf{K}_1-\mathbf{K}}_{n_3,n_4,0}\langle B^\dagger_{\nu_4}B^\dagger_{\nu_3}B_{\nu_1}B_\mu\rangle - \widetilde{W}^{n_1,n_3,\mathbf{K}_1-\mathbf{K}'}_{m,n_4,0}\langle B^\dagger_\nu B^\dagger_{\nu_4}B_{\nu_3}B_{\nu_1}\rangle\}+\mathcal{O}(\mathcal{R}^6), \tag{29}$$

where $\omega^0_{\nu\mu}\equiv\omega^0_\nu-\omega^0_\mu$, $T_{\nu\mu}=T^{\rm intra}_2$, $\nu=\{n,\mathbf{K}\}$, and $\mu=\{m,\mathbf{K}'\}$. The expectation values of higher powers of the exciton operators can be derived in a similar manner. For the interband-intraband transition

$$\left(i\frac{d}{dt}+\omega^0_{43;2}+\frac{i}{T_{43;2}}\right)\langle B^\dagger_{\nu_4}B^\dagger_{\nu_3}B_{\mu_2}\rangle = \mathcal{R}_{\nu_4}\langle B^\dagger_{\nu_3}B_{\nu_2}\rangle+\mathcal{R}_{\nu 3}\langle B^\dagger_{\nu_4}B_{\nu_2}\rangle-\mathcal{R}^*_{\nu_2}\langle B^\dagger_{\nu_4}B^\dagger_{\nu_3}\rangle + \mathcal{O}(\mathcal{R}^5), \tag{30}$$

with $\omega^0_{43;2}\equiv\omega^0_{\nu_4}+\omega^0_{\nu_3}-\omega^0_{\nu_2}$, while, for the biexciton operator,

$$\left(i\frac{d}{dt}+\omega^0_{\nu_4\nu_1}+\omega^0_{\nu_3\nu_2}+\frac{i}{T_{43;21}}\right)\langle B^\dagger_{\nu_4}B^\dagger_{\nu_3}B_{\nu_2}B_{\nu_1}\rangle = \mathcal{R}_{\nu_4}\langle B^\dagger_{\nu_3}B_{\nu_2}B_{\nu_1}\rangle+\mathcal{R}_{\nu_3}\langle B^\dagger_{\nu_4}B_{\nu_2}B_{\nu_1}\rangle-\mathcal{R}^*_{\nu_2}\langle B^\dagger_{\nu_4}B^\dagger_{\nu_3}B_{\nu_1}\rangle - \mathcal{R}^*_{\nu_1}\langle B^\dagger_{\nu_4}B^\dagger_{\nu_3}B_{\nu_2}\rangle+\mathcal{O}(\mathcal{R}^6). \tag{31}$$

The decay times have been written with a subscript that allows them to vary with the mode under consideration, for example to allow the diagonal terms in Eq. (29) to decay more slowly than the off-diagonal terms. Once eigenvectors of H_0 are known analytically or numerically in $\mathbf{k}$ space, χ and $\widetilde{W}$ can be found, and calculations to order $\mathcal{R}^4$ can be performed using Eqs. (28)–(31) or to higher order in the optical field by deriving a more general set of equations using Eq. (23).

The solution to each of Eqs. (28)–(31) consists of a homogeneous term which is a damped oscillation the natural frequency of the mode plus a coherently driven term. The equation

$$(id/dt+\omega)\langle A\rangle=-\mathcal{F}$$

has the solution

$$\langle A\rangle=\left\{\langle A^S\rangle+i\int_{-\infty}^{t}dt'e^{-i\omega t'}\mathcal{F}(t')\right\}e^{i\omega t},$$

where A^S is the time-independent Schrödinger operator. The coherent part goes as a power of the field though $\mathcal{R}_\nu$, and the equations can be solved to any specified order in the driving electric field. The iterative solution for these expectation values can be written down explicitly as an expansion in the driving field. An expression of the form $\langle B^{\dagger m}B^{n-m}\rangle$ includes terms up to order n in the field, as can be verified by noting from Eq. (28) that $\langle B\rangle$ is of order $\mathcal{R}$ and higher, and from Eqs. (29)–(31) that the order in the field goes up as n. The interband functions are

$$\langle B_\nu^\dagger\rangle_t=\left\{\langle B_\nu^{S\dagger}\rangle+i\int_{-\infty}^{t}dt'e^{-i\omega_\nu t'}\mathcal{R}_\nu(t')\right\}e^{i\omega_\nu t}+\mathcal{O}(\mathcal{R}^3).$$

For the expectation values of the intraband operators, this gives

$$\langle B_\nu^\dagger B_\mu\rangle_t=\left\{\langle B_\nu^{S\dagger}B_\mu^S\rangle+i\int_{-\infty}^{t}dt'e^{-i\omega_{\nu\mu}t'}[\mathcal{R}_\nu(t')\langle B_\mu\rangle_{t'}-\mathcal{R}_\mu^*(t')\langle B_\nu^\dagger\rangle_{t'}]\right\}e^{i\omega_{\nu\mu}t}+\mathcal{O}(\mathcal{R}^4),\tag{32}$$

and, for the interband-intraband operators,

$$\langle B_{\nu_4}^\dagger B_{\nu_3}^\dagger B_{\nu_2}\rangle_t=\left\{\langle B_{\nu_4}^{S\dagger}B_{\nu_3}^{S\dagger}B_{\nu_2}^S\rangle+i\int_{-\infty}^{t}dt'e^{-i\omega_{43;2}t'}[\mathcal{R}_{\nu_4}(t')\langle B_{\nu_3}^\dagger B_{\nu_2}\rangle_{t'}+\mathcal{R}_{\nu3}(t')\langle B_{\nu_4}^\dagger B_{\nu_2}\rangle_{t'}-\mathcal{R}_{\nu_2}^*(t')\langle B_{\nu_4}^\dagger B_{\nu_3}^\dagger\rangle_{t'}]\right\}\times e^{i\omega_{43;2}t}+\mathcal{O}(\mathcal{R}^5).\tag{33}$$

In these expressions, $\omega\equiv\omega^0+i/T$ incorporates the dephasing time constants.

If damping is omitted, there are simple relationships among the expectation values of products of the interband and intraband operators. This follows from the fact that they are all just expectation values of powers of the same interband annihilation and creation operators. The operator equation (23) can be solved for the operator $B_\nu^\dagger$, to obtain

$$B_\nu^\dagger(t)=(B_\nu^{S\dagger}+iY_\nu)e^{i\omega_\nu^0 t}+\mathcal{O}(Y^3),\tag{34}$$

where

$$Y_\nu(t)\equiv\int_{-\infty}^{t}dt'\mathcal{R}_\nu e^{-i\omega_\nu^0 t'}.\tag{35}$$

For simplicity, we will assume here that $H_{\rm int}$ includes only incoherent effects so that only Schrödinger number operator expectation values of the form $\langle B_\nu^{S\dagger}B_\nu^S\rangle$ are nonzero. These terms count excitons incoherently scattered and dephased. Then

$$\langle B_\nu^\dagger B_\mu\rangle_t^{T=\infty}=(Y_\nu Y_\mu^*+\langle B_\nu^{S\dagger}B_\mu^S\rangle)e^{i\omega_{\nu\mu}^0 t}+\mathcal{O}(Y^4)\tag{36}$$

and

$$\langle B_{\nu_4}^\dagger B_{\nu_3}^\dagger B_{\nu_2}\rangle_t^{T=\infty}=(iY_{\nu_4}Y_{\nu_3}Y_{\nu_2}^*+iY_{\nu_4}\delta_{\nu_3,\nu_2}\langle B_{\nu_3}^{S\dagger}B_{\nu_2}^S\rangle+iY_{\nu_3}\delta_{\nu_4,\nu_2}\langle B_{\nu_4}^{S\dagger}B_{\nu_2}^S\rangle)e^{i\omega_{43;2}^0 t}+\mathcal{O}(Y^5).\tag{37}$$

Deviations from these expressions are due to different time constants, which modify $Y_\nu(t)$ due to decay of the lower-order modes that are driving it. If all the time constants are the same, this effect drops out due to cancellation of the time constants in integrals such as Eqs. (32) and (33), and they become just the undamped functions (36) and (37) multiplied by e^{-t/T_2}. According to the above equations, it is never strictly correct to multiply the damped functions together. However, since we are using an exciton basis here, the electron and hole cannot decouple for any treatment of the time constants.

V. DISCUSSION OF SBE'S AND DCT, AND SUMMARY

The unique feature of the present description of the dynamics of the semiconductor band edge is that it is based on exciton operators in an ordered pair space while including phase-space filling. Hamiltonians (13)–(16), except for the inclusion of coupling to the field, are essentially identical to the result obtained by Hanamura and Haug[6] for strictly bosonic excitons. However, the dynamical operator equation (23) that we derive from this Hamiltonian utilizes the quasibosonic commutation relations (11) and thus incorporates phase-space-filling effects. The expansion in powers of the field used here is analogous to the DCT scheme of Axt and Stahl.[2,3] Here the use of ordered electron-hole pairs in a qboson space simplifies the expansion in powers of the field for optically created excitons. The expectation values of the coherent interband response and the number operators without damping give the SBE. We will now consider these points in more detail.

An analogy can be drawn between Eqs. (28)–(31) and DCT. In contrast to the present work, Axt and Stahl use a basis of electron-hole pairs created at a fixed lattice sites, except for Ref. 3 where they work in $\mathbf{k}$ space. However, the equations that they derive are very similar in form to Eqs. (28)–(31). The exciton expectation values $\langle B_\nu^\dagger\rangle$, $\langle B_\nu^\dagger B_\mu\rangle$,

$\langle B^{\dagger}_{\nu_4}B^{\dagger}_{\nu_3}B_{\nu_2}\rangle$, $\langle B^{\dagger}_{\nu_4}B^{\dagger}_{\nu_3}B_{\nu_2}B_{\nu_1}\rangle$, and $\langle B_{\nu}B_{\mu}\rangle$ in our formalism correspond to Axt and Stahl's electron-hole expectation values Y^*, $\{N,C,D,B\}$, $\{S^*,T,Z^*\}$, $\{K,L\}$, and B, respectively. With this substitution, the form of our equations agrees term by term with those in Ref. 2. They also conclude that deviations from $N=Y\times Y$ to order $\mathcal{E}^4$ are a consequence of differences in damping rates.[3] Our result does not imply the relationships among these damping rates discussed in Ref. 8.

If the ω^0 terms in Eqs. (28) and (29) are transformed back to $\mathbf{k}$ space as discussed in the last paragraph of Sec. III, and $\psi_{n,\mathbf{k}}=\delta_{\mathbf{k}_n,\mathbf{k}}$ is substituted into Eqs. (25) and (26), one obtains

$$\left(i\hbar\frac{d}{dt}+E^0_k+\frac{i\hbar}{T_2^{\text{inter}}}\right)\langle B^{\dagger}_{\mathbf{k}}\rangle$$
$$=\mathcal{E}(t)d_{\text{cv}}(1-2\langle B^{\dagger}_{\mathbf{k}}B_{\mathbf{k}}\rangle)+\sum_{\mathbf{q}\neq 0}V_q(\langle B^{\dagger}_{\mathbf{k}+\mathbf{q}}\rangle$$
$$-2\langle B^{\dagger}_{\mathbf{k}+\mathbf{q}}B^{\dagger}_{\mathbf{k}}B_{\mathbf{k}}\rangle)+2\sum_{\mathbf{q}\neq 0}V_q\langle B^{\dagger}_{\mathbf{k}}B^{\dagger}_{\mathbf{k}+\mathbf{q}}B_{\mathbf{k}+\mathbf{q}}\rangle$$
$$+\mathcal{O}(\mathcal{R}^5) \tag{38}$$

and

$$\left(i\hbar\frac{d}{dt}+\frac{i\hbar}{T_2^{\text{intra}}}\right)\langle B^{\dagger}_{\mathbf{k}}B_{\mathbf{k}}\rangle$$
$$=\mathcal{E}(t)d_{\text{cv}}\langle B_{\mathbf{k}}\rangle-\mathcal{E}(t)d^*_{\text{cv}}\langle B_{\mathbf{k}}\rangle^*+2\sum_{\mathbf{q}\neq 0}V_q\langle B^{\dagger}_{\mathbf{k}+\mathbf{q}}B_{\mathbf{k}}\rangle$$
$$-2\sum_{\mathbf{q}\neq 0}V_q\langle B^{\dagger}_{\mathbf{k}}B_{\mathbf{k}-\mathbf{q}}\rangle+\mathcal{O}(\mathcal{R}^4). \tag{39}$$

Except for omission of the static field, Eq. (38) and the linear approximation to Eq. (39) are identical to the DCT equations of Ref. 3. Without damping, $B^{\dagger}_{\mathbf{k}}(t)=\Sigma_{\nu}\psi^*_{n,\mathbf{k}}(B^{S\dagger}_{\mathbf{n}}+iY_{\nu})e^{i\omega^0_{\nu}t}$ can be obtained from Eqs. (34) and (9). This implies that $\langle B^{\dagger}_{\mathbf{k}+\mathbf{q}}B^{\dagger}_{\mathbf{k}}B_{\mathbf{k}}\rangle$ and $\langle B^{\dagger}_{\mathbf{k}}B^{\dagger}_{\mathbf{k}+\mathbf{q}}B_{\mathbf{k}+\mathbf{q}}\rangle$ can be written as a sum of terms of the form (37) factored into polarization and number operator parts using Eq. (36) to give $\langle B^{\dagger}_{\mathbf{k}+\mathbf{q}}\rangle\times\langle B^{\dagger}_{\mathbf{k}}B_{\mathbf{k}}\rangle$ and $\langle B^{\dagger}_{\mathbf{k}}\rangle\langle B^{\dagger}_{\mathbf{k}+\mathbf{q}}B_{\mathbf{k}+\mathbf{q}}\rangle$, respectively. Equations (38) and (39) then reduce to the SBE in the zero damping, zero center-of-mass motion limit.

To summarize, we developed a theory of quasibosonic excitons that is based on use of exciton creation operators to describe interband and intraband polarization and higher-order effects. It is consistent with DCT's and reduces to the SBE when terminated at fourth order in the electric field if differences in relaxation times are ignored. In our theory electron-hole pairs are to be thought of as qbosons that satisfy commutation relations intermediate between bosonic and fermionic. These pairs can be transformed to a basis of bound and unbound excitons that incorporates the intrapair Coulomb interaction. All exciton-exciton interactions, including exchange, are included in a single interaction energy that depends only on their center-of-mass momenta and hydrogenic state. These excitons are the normal modes of the system, and their energies and energy differences determine the frequencies that characterize its optical response. This qboson picture of excitons leads us to a simplified description of the physics of optical processes of semiconductors near the band edge. Quantitative predictions of the theory can be obtained using Eqs. (28)–(31). Equations that are valid to higher order in the optical field can be derived using (23).

Only the interband exciton creation operator and its adjoint, the destruction operator B_{ν}, are required. Products of these operators can then be formed that describe intraband transitions, exciton numbers, and higher-order processes. Since the driving force is an electric field that does not involve the exciton operator (and is just a function unless the field is second quantized), the coherent response is also just a function that can be taken outside the expectation value. The homogeneous solution which is the Schrödinger operator multiplied by $e^{i\omega t}$ normally only plays a role for the counting operators that describe incoherent processes. If damping is neglected, the operators describing interband and interband transitions and numbers are simply related since they are just expectation values of products of the exciton operators and the coherent response is just a (nonoperator) function of time.

When making comparisons with experiment, dephasing and the consequent damping of coherent oscillations cannot be ignored. Since these effects are described by decay of the off-diagonal elements of the system's density matrix, they are not part of the Heisenberg operator dynamics, and they modify these simple relationships. The SBE's rely on these relationships, and can produce unphysical results if the interband polarization that drives the Coulomb coupling of the electron and hole decays faster than the intraband polarization,[3,9] since factorization of the damped functions implies that Coulomb coupling will decay twice as fast as interband polarization. DCT and the present work predict that the electron and hole will remain coupled as observed.[4] We conclude that the relationship $T_2^{\text{intra}}=T_2^{\text{inter}}/2$ has no rigorous theoretical basis, and in fact it is $T_2^{\text{intra}}=T_2^{\text{inter}}$ that results in simple relationships among the interband, intraband, and interband-interband processes. In this case the damping can be added simply by multiplying all the undamped expectation values by e^{-t/T_2}.

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APPENDIX: TRANSFORMATION TO PAIR SPACE

Here the fermion operator $A_F=\alpha^{\dagger}_{\mathbf{k}}\alpha_{\mathbf{k}'}$ will be transformed to pair space using Eqs. (4) and (5), and then other operators will be considered. Only the Nth term in the expansion of the exponential acts on a state containing N pairs, since higher powers of b give zero and lower powers are eliminated by the projection operator P_F. All of the fermion operators commute with all of the qboson operators. When acting on a state vector containing N pairs, the qboson operator is

$$(\alpha^{\dagger}_{\mathbf{k}}\alpha_{\mathbf{k}'})_B=\frac{1}{N!^2}O\sum_{k_i,k_i'} B^{\dagger}_{k_1}\ldots B^{\dagger}_{k_N}B_{k_N'}\ldots B_{k_1'}\beta_{\mathbf{k}_{h_1}} \times\alpha_{\mathbf{k}_{e_1}}\ldots\beta_{\mathbf{k}_{h_N}}\alpha_{\mathbf{k}_{e_N}}\alpha^{\dagger}_{\mathbf{k}}\alpha_{\mathbf{k}'}\alpha^{\dagger}_{\mathbf{k}_{e_N}'}\beta^{\dagger}_{\mathbf{k}_{h_N}'}\ldots \times\alpha^{\dagger}_{\mathbf{k}_{e_1}'}\beta^{\dagger}_{\mathbf{k}_{h_1}'}.$$

An even number of exchanges move all electron operators to the right of all hole operators. Since there are no electrons or holes in the initial qboson state, only those electrons and holes that are created can subsequently be destroyed. They can be destroyed in any order, and the sign will depend on the number of exchanges of the wave vectors. For the electrons, $\mathbf{k}$ and $\mathbf{k}'$ can also be permuted. Operators P_e and P_h will be defined that generate all permutations of the electron and hole wave vectors, and give a factor $+1$ if there is an even number of exchanges, and -1 if there is an odd number. Then

$$(\alpha^{\dagger}_{\mathbf{k}}\alpha_{\mathbf{k}'})_B=\frac{1}{N!^2}O\sum_{k_i} P_eP_hB^{\dagger}_{k_1}\ldots B^{\dagger}_{\mathbf{k},\mathbf{k}_{h_N}}B_{\mathbf{k}',\mathbf{k}_{hN}}\ldots B_{k_1}.$$

The operators are symmetric under exchange of qbosons, and thus electrons and holes need not both be exchanged. We will permute only electrons and pairs. There are $N!$ equivalent pairs and the ordering operator eliminates permutations where pairing has been changed. For a state containing n pairs, $\Sigma_k B^{\dagger}_k B_k=n$, where $n=1$ for the innermost factor to $N-1$ giving $(N-1)!$. There are N holes that $\mathbf{k}'$ can be paired up with, so we obtain

$$(\alpha^{\dagger}_{\mathbf{k}}\alpha_{\mathbf{k}'})_B=\sum_{\mathbf{k}_h} B^{\dagger}_{\mathbf{k},\mathbf{k}_h}B_{\mathbf{k}',\mathbf{k}_h}, \tag{A1}$$

which is independent of N and hence is the qboson operator for an arbitrary state. The operator $\beta^{\dagger}_{\mathbf{k}}\beta_{\mathbf{k}'}$ is transformed similarly, with the roles of electrons and holes reversed so that

$$(\beta^{\dagger}_{\mathbf{k}}\beta_{\mathbf{k}'})_B=\sum_{\mathbf{k}_e} B^{\dagger}_{\mathbf{k}_e,\mathbf{k}}B_{\mathbf{k}_e,\mathbf{k}'}. \tag{A2}$$

The above-transformed operators give the single-exciton contribution to the Hamiltonian and the polarization. For the commutator there is an additional requirement of consistency with its definition $B_{k_1}B^{\dagger}_{k_2}-B^{\dagger}_{k_2}B_{k_1}$. The qboson commutation relations obtained by substitution in Eq. (3) are

$$[B_{k_1},B^{\dagger}_{k_2}]=\delta_{k_1,k_2}-\delta_{\mathbf{k}_{h1},\mathbf{k}_{h2}}\sum_{\mathbf{k}_h} B^{\dagger}_{\mathbf{k}_{e2},\mathbf{k}_h}B_{\mathbf{k}_{e1},\mathbf{k}_h} - \delta_{\mathbf{k}e_1,\mathbf{k}e_2}\sum_{\mathbf{k}_e} B^{\dagger}_{\mathbf{k}_e,\mathbf{k}_{h2}}B_{\mathbf{k}_e,\mathbf{k}_{h1}}.$$

There appears to be a problem with this equation, since the left-hand side gives zero unless the pair k_1 is present, while the right-hand side can be nonzero due to the sums over the dummy variables $\mathbf{k}_h$ and $\mathbf{k}_e$. To resolve this apparent contradiction, we will consider the effect of the above commutation relation on a state the contains a hole $\mathbf{k}_{h1}$ and electron $\mathbf{k}_e$, that is, the state $|\mathbf{k}_e,\mathbf{k}_{h_1};\ldots\rangle$. Consider $\mathbf{k}_{h_1}\neq\mathbf{k}_{h_2}$ so that only the last term contributes on the right-hand side. If $\mathbf{k}_e=\mathbf{k}_{e_1}$, then the left- and right-hand sides agree. If $\mathbf{k}_e\neq\mathbf{k}_{e_1}$ the last term on the right gives $-\delta_{\mathbf{k}e_1,\mathbf{k}e_2}|\mathbf{k}_e,\mathbf{k}_{h_2};\ldots\rangle$ while the $B_{\mathbf{k}_{e1},\mathbf{k}_{h1}}B^{\dagger}_{\mathbf{k}e_2,\mathbf{k}_{h2}}$ on its left-hand side gives $B_{\mathbf{k}_{e1},\mathbf{k}_{h1}}|\mathbf{k}_{e_2},\mathbf{k}_{h_2};\mathbf{k}_e,\mathbf{k}_{h_1};\ldots\rangle=0$. The apparent inconsistency arises because the fermion operators know nothing of pairing. If exchange of $\mathbf{k}_e$ and $\mathbf{k}_{e_2}$ were to be allowed, the left-hand side would become $-\delta_{\mathbf{k}e_1,\mathbf{k}e_2}|\mathbf{k}_e,\mathbf{k}_{h_2};\ldots\rangle$, in agreement with the right-hand side. The ordering prescription thus eliminates $\mathbf{k}_e\neq\mathbf{k}_{e_1}$ (and $\mathbf{k}_h\neq\mathbf{k}_{h_1}$ by an analogous argument) and reduces the commutation relation to Eq. (7).

The Hamiltonian can be transformed in an similar manner to $\alpha^{\dagger}_{\mathbf{k}}\alpha_{\mathbf{k}'}$. This is straightforward for H_0, which is a special case of Eqs. (A1) and (A2), and for H_I. For the two-pair electron-electron Coulomb interaction

$$(\alpha^{\dagger}_{\mathbf{k}_1-\mathbf{q}}\alpha^{\dagger}_{\mathbf{k}_2+\mathbf{q}}\alpha_{\mathbf{k}_2}\alpha_{\mathbf{k}_1})_B =\frac{1}{N!}O\sum_{k_i} P_eB^{\dagger}_{\mathbf{k}_2+\mathbf{q},\mathbf{k}_{h_N}}B^{\dagger}_{\mathbf{k}_1-\mathbf{q},\mathbf{k}_{h_{N-1}}}\ldots B^{\dagger}_{k_1} \times B_{k_1}\ldots B_{\mathbf{k}_1,\mathbf{k}_{h_{N-1}}}B_{\mathbf{k}_2,\mathbf{k}_{h_N}},$$

which reduces to

$$(\alpha^{\dagger}_{\mathbf{k}_1-\mathbf{q}}\alpha^{\dagger}_{\mathbf{k}_2+\mathbf{q}}\alpha_{\mathbf{k}_2}\alpha_{\mathbf{k}_1})_B =\sum_{\mathbf{k}_{h_1},\mathbf{k}_{h_2}}(B^{\dagger}_{\mathbf{k}_1-\mathbf{q},\mathbf{k}_{h_1}}B^{\dagger}_{\mathbf{k}_2+\mathbf{q},\mathbf{k}_{h_2}}B_{\mathbf{k}_2,\mathbf{k}_{h_2}}B_{\mathbf{k}_1,\mathbf{k}_{h_1}} -B^{\dagger}_{\mathbf{k}_2+\mathbf{q},\mathbf{k}_{h_1}}B^{\dagger}_{\mathbf{k}_1-\mathbf{q},\mathbf{k}_{h_2}}B_{\mathbf{k}_2,\mathbf{k}_{h_2}}B_{\mathbf{k}_1,\mathbf{k}_{h_1}}).$$

For the electron-hole interactions,

$$(\alpha^{\dagger}_{\mathbf{k}_1-\mathbf{q}}\beta^{\dagger}_{\mathbf{k}_2+\mathbf{q}}\beta_{\mathbf{k}_2}\alpha_{\mathbf{k}_1})_B =\frac{1}{N!}O\sum_{k_i} P_eB^{\dagger}_{\mathbf{k}_2+\mathbf{q},\mathbf{k}_{h_1}}B^{\dagger}_{\mathbf{k}_{e_1},\mathbf{k}_1-\mathbf{q}}\ldots B^{\dagger}_{k_1}B_{k_1}\ldots \times B_{\mathbf{k}_{e_1},\mathbf{k}_1}B_{\mathbf{k}_2,\mathbf{k}_{h_1}}$$

becomes

$$(\alpha^{\dagger}_{\mathbf{k}_1-\mathbf{q}}\beta^{\dagger}_{\mathbf{k}_2+\mathbf{q}}\beta_{\mathbf{k}_2}\alpha_{\mathbf{k}_1})_B =\sum_{k_1}(B^{\dagger}_{\mathbf{k}_{e_1},\mathbf{k}_1-\mathbf{q}}B^{\dagger}_{\mathbf{k}_2+\mathbf{q},\mathbf{k}_{h_1}}B_{\mathbf{k}_2,\mathbf{k}_{h_1}}B_{\mathbf{k}_{e_1},\mathbf{k}_1} -B^{\dagger}_{\mathbf{k}_{e_1},\mathbf{k}_1-\mathbf{q}}B^{\dagger}_{\mathbf{k}_2+\mathbf{q},\mathbf{k}_{h_1}}B_{\mathbf{k}_{e_1},\mathbf{k}_{h_1}}B_{\mathbf{k}_2,\mathbf{k}_1} -B^{\dagger}_{\mathbf{k}_2+\mathbf{q},\mathbf{k}_1-\mathbf{q}}B^{\dagger}_{\mathbf{k}_{e_1},\mathbf{k}_{h_1}}B_{\mathbf{k}_2,\mathbf{k}_{h_1}}B_{\mathbf{k}_{e_1},\mathbf{k}_1}) +B^{\dagger}_{\mathbf{k}_2+\mathbf{q},\mathbf{k}_1-\mathbf{q}}B_{\mathbf{k}_2,\mathbf{k}_1}.$$

This gives Eqs. (14)–(16) after summation over $\mathbf{k}_1$ and $\mathbf{k}_2$, and conversion to center-of-mass and relative coordinates.

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