

Generalized semiconductor Bloch equations: Local fields and transient gratings

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Equations of motion for two-point equal-time electronic correlation functions, which describe near-resonant optical excitation of semiconductors in the Hartree approximation, are derived. These equations generalize the common semiconductor Bloch equations to incorporate inhomogeneous excitation of the medium and strong dielectric effects. We show that the many-body effects on the optical non-linearity cannot be fully included by simply replacing the external electric field by the local field.

I. INTRODUCTION

The properties of semiconductors excited by laser light are currently attracting considerable interest.¹⁻⁶ Early investigations primarily focused on the theory of the many-body effects related to interacting electrons and holes.^{7,8} Later this area acquired a growing attention triggered by experiments performed using picosecond and femtosecond laser pulses. Early experiments of this type investigated the Stark shift of the excitonic lines^{9,10} and the reduction of oscillator strength of the excitonic medium induced by optical excitation.¹¹⁻¹³ Numerous nonlinear optical effects in semiconductors were subsequently investigated, including phonon-induced non-linearity and the time-dependent optical Stark effect.¹⁴⁻²⁷ Particular attention was devoted to the effect of optical saturation in semiconductor nanostructures with reduced dimensionality using an excitation quasi-resonant with an excitonic line.^{1,3,11-16,19-21} In this case, optical excitation can be attributed to virtual excitonic population, and the nonlinear excitonic effect can be described in terms of the Stark shift of the excitonic line and the renormalization of the oscillator strength (phase-space filling). These effects have been widely explored experimentally^{1,9,11-13} and the results were found to be in very good agreement with theoretical calculations performed using the Keldysh nonequilibrium diagrammatic technique.^{1,2} When the interaction of carriers with phonons is neglected (which is always justified for off-resonant excitation), and neglecting the hole band anisotropy and the exchange interaction, these theories result in the semiconductor Bloch equations (SBE).¹ These equations are commonly used to describe the optical properties of semiconductors driven by external electromagnetic fields, and are completely analogous to the Bloch equations used for atomic excitation. The main approximation in deriving the SBE is the Hartree factorization of higher electronic correlation functions. Additional important approximations are the assumption of uniform excitation of the electronic medium, which implies that two-point electronic correlation functions depend only on the relative variable and not on the center-of-mass variable (i.e., translational invariance), and the neglect of the dipole-dipole interaction between unit cells of the semiconductor. The uniform approximation for the excitation is usually valid for pump-

probe experiments, and the neglect of dipole-dipole interactions (the external field approximation) may be justified for optically thin systems (quantum wells and quantum dots). The SBE were found to be adequate for the description of the Stark shift and phase-space filling in semiconductor quantum wells.¹¹⁻¹³

In this paper we derive the equations of motion for second-order spatial (equal-time) electronic correlation functions of semiconductors, using the Hartree factorization. We consider a general nonuniform excitation and take into account the dipole-dipole interaction between unit cells (see Sec. II). Nonuniform excitation clearly needs to be considered in grating experiments,²⁸ where the density of carriers varies on a length scale comparable to the optical wavelength. The dipole-dipole interaction is responsible for the strong dielectric effect in semiconductors. This interaction is particularly important for bulk semiconductors and potentially also for large semiconductor particles and thick semiconductor layers. The generalized semiconductor Bloch equations (GSBE) for electronic correlation functions derived in this paper extend the standard semiconductor Bloch equations to include nonuniform optical excitations and strong dielectric effects. In the limit of uniform excitation, the equations can be recast in the local-field form. We show, however, that it is impossible to incorporate all the effects of dipole-dipole interaction in the local field, and additional contributions to the optical nonlinearity coming from the dipole-dipole interaction between unit cells need to be considered. When the dipole-dipole interaction is neglected and the excitation is taken to be uniform, we recover the conventional SBE.¹

II. GENERALIZED SEMICONDUCTOR BLOCH EQUATIONS

The minimal coupling ($\mathbf{p} \cdot \mathbf{A}$) Hamiltonian provides an exact and a commonly used starting point for the microscopic theory of electronic systems coupled with the radiation field.²⁹ For systems with localized electronic states (e.g., molecular crystals), a canonical transformation can bring the Hamiltonian to the multipolar ($\boldsymbol{\mu} \cdot \mathbf{D}$) in which the local-field approximation and the connection with the Bloch equations is more transparent.^{29,30} When the transverse electric field is treated classically, that goal

may be accomplished using a similar procedure based on a gauge transformation, which brings the transverse field explicitly into the Hamiltonian. This transformation, which does not assume localized electronic states, and can therefore be applied to molecular as well as semiconductor systems, is carried out in Appendix A, resulting in the following two-band model Hamiltonian of a semiconductor:⁶

$$H = \int d\mathbf{r} \hat{\psi}^\dagger(\mathbf{r}) \left[\frac{\hat{p}^2}{2m} + V_B(\mathbf{r}) + e\mathbf{r} \cdot \mathbf{E}_T(\mathbf{r}, t) \right] \hat{\psi}(\mathbf{r}) + \int d\mathbf{r}_1 d\mathbf{r}_2 \hat{\psi}^\dagger(\mathbf{r}_1) \hat{\psi}^\dagger(\mathbf{r}_2) \hat{\psi}(\mathbf{r}_2) \hat{\psi}(\mathbf{r}_1) V_c(\mathbf{r}_1 - \mathbf{r}_2). \quad (2.1)$$

Here, $\hat{\psi}(\mathbf{r})$ is the electronic annihilation operator at point \mathbf{r} , $\hat{p}(\mathbf{r})$ is the electron momentum operator, $V_B(\mathbf{r})$ is the periodic lattice potential, $V_c(\mathbf{r})$ is the (instantaneous) Coulomb potential, and $\mathbf{E}_T(\mathbf{r})$ is the classical transverse electric field (we use the Coulomb gauge). The electronic field operator written using the Bloch state basis set has the form

$$\hat{\psi}(\mathbf{r}) = \sum_{\mathbf{k}} [\hat{c}_{\mathbf{k}} \phi_{c\mathbf{k}}(\mathbf{r}) + \hat{d}_{\mathbf{k}}^\dagger \phi_{v\mathbf{k}}(\mathbf{r})], \quad (2.2)$$

where $\hat{c}_{\mathbf{k}}$ ($\hat{d}_{\mathbf{k}}$) denotes an annihilation operator for an electron (hole) with momentum \mathbf{k} ($-\mathbf{k}$). These operators satisfy the Fermi anticommutation rules $\{\hat{c}_{\mathbf{k}}, \hat{c}_{\mathbf{k}'}^\dagger\} = 2\delta_{\mathbf{k}\mathbf{k}'}$ and $\{\hat{d}_{\mathbf{k}}, \hat{d}_{\mathbf{k}'}^\dagger\} = 2\delta_{\mathbf{k}\mathbf{k}'}$. $\phi_{c\mathbf{k}}(\mathbf{r})$ and $\phi_{v\mathbf{k}}(\mathbf{r})$ are the corresponding Bloch functions. The model of the electronic system given by Eqs. (2.1) and (2.2) contains several approximations. First, the spin index has been omitted in the electronic variables, which means that we neglect the Hubbard-type Coulomb-induced coupling between states with different spins. We have further neglected the degeneracy of the hole band, i.e., the heavy- and light-hole effect. Both approximations enter in the same way, as discussed in Ref. 1. In Appendix B we substitute Eq. (2.2) in Eq. (2.1) and, after making a series of approximations that will be discussed below, we obtain

$$H = H_1 + H_2$$

where

$$H_1 = \sum_{\mathbf{k}} [\varepsilon_c(\mathbf{k}) \hat{c}_{\mathbf{k}}^\dagger \hat{c}_{\mathbf{k}} + \varepsilon_v(\mathbf{k}) \hat{d}_{\mathbf{k}}^\dagger \hat{d}_{\mathbf{k}}] + \sum_{\mathbf{k}q} \bar{d}_{cv}[\mathbf{E}_T(\mathbf{q}, t) \hat{c}_{\mathbf{k}+q}^\dagger \hat{d}_{-\mathbf{k}} + \text{H.c.}] + \sum_{\mathbf{k}, \mathbf{k}'} \mathbf{E}_T(\mathbf{k} - \mathbf{k}', t) [\bar{d}_{cc}(\mathbf{k}, \mathbf{k}') \hat{c}_{\mathbf{k}}^\dagger \hat{c}_{\mathbf{k}'} + \bar{d}_{vv}(\mathbf{k}, \mathbf{k}') \hat{d}_{\mathbf{k}}^\dagger \hat{d}_{\mathbf{k}'}], \quad (2.3a)$$

$$H_2 = \frac{1}{2} \sum_{\mathbf{k}_1, \mathbf{k}_2, q} [\bar{V}_q (\hat{c}_{\mathbf{k}_1}^\dagger \hat{c}_{\mathbf{k}_2}^\dagger \hat{c}_{\mathbf{k}_2+q} \hat{c}_{\mathbf{k}_1-q} + \hat{d}_{\mathbf{k}_1}^\dagger \hat{d}_{\mathbf{k}_2}^\dagger \hat{d}_{\mathbf{k}_2+q} \hat{d}_{\mathbf{k}_1-q}) - 2(\bar{V}_q + \bar{V}_q) \hat{c}_{\mathbf{k}_1}^\dagger \hat{d}_{\mathbf{k}_2}^\dagger \hat{d}_{\mathbf{k}_2+q} \hat{c}_{\mathbf{k}_1-q}]. \quad (2.3b)$$

Here, H_1 and H_2 denote the single-particle and the two-particle parts of the Hamiltonian. \bar{d} are the transition dipole matrix elements, both intraband ($\bar{d}_{cc}, \bar{d}_{vv}$) and interband (\bar{d}_{cv}). The Fourier transforms of the monopole-monopole and dipole-dipole parts of the Coulomb poten-

tial are

$$\bar{V}_q = \sum_n e^{iq \cdot \mathbf{R}_n} \frac{e^2}{|\mathbf{R}_n|}, \quad (2.3c)$$

$$\bar{V}_q = \sum_n e^{iq \cdot \mathbf{R}_n} \frac{2e^2 \bar{d}_{cv}^2}{|\mathbf{R}_n|^3}, \quad (2.3d)$$

and $\varepsilon_c(\mathbf{k}) \equiv E_c(\mathbf{k})$ and $\varepsilon_v(\mathbf{k}) \equiv E_v(\mathbf{k}) + \sum_{q \neq 0} (\bar{V}_q + \frac{1}{2} \bar{V}_q)$, where $E_v(\mathbf{k})$ and $E_c(\mathbf{k})$ are the single-particle energies of the valence and conduction bands, respectively. In the derivation of Eqs. (2.3) we have performed a multipolar expansion of the electron-electron interaction between cells and retained only the monopole-monopole and dipole-dipole terms. We further imposed a normal ordering with respect to electrons and holes and neglected terms with the reverse ordering in order to define properly the vacuum (ground state). In addition, we have assumed the electric field to be uniform over a unit cell (see Appendix A), and the Bloch functions have been approximated as follows:

$$\phi_{c\mathbf{k}}(\boldsymbol{\rho} + \mathbf{R}_n) = e^{i\mathbf{k} \cdot (\boldsymbol{\rho} + \mathbf{R}_n)} u_{c\mathbf{k}}(\boldsymbol{\rho}) \equiv e^{i\mathbf{k} \cdot \mathbf{R}_n} u_{c\mathbf{k}_0}(\boldsymbol{\rho}), \quad (2.4)$$

where \mathbf{k}_0 denotes the wave vector at the center of the Brillouin zone and \mathbf{R}_n is the center of the n th unit cell so that $\mathbf{r} = \mathbf{R}_n + \boldsymbol{\rho}$. We have thus neglected the momentum variation of the periodic part (u_c) of the Bloch function. A similar approximation was made for the valence band involving ϕ_v and u_v . With these approximations the interband and dipole intraband matrix elements are given by

$$\bar{d}_{cv} = \int_{-R_c/2}^{R_c/2} u_c(\boldsymbol{\rho}) u_v(\boldsymbol{\rho}) \boldsymbol{\rho} d\boldsymbol{\rho}, \quad (2.5a)$$

$$\bar{d}_{cc(vv)}(\mathbf{k}_1, \mathbf{k}_2) = \sum_{\mathbf{R}_n} \mathbf{R}_n \int d\boldsymbol{\rho} u_{c(v)}(\boldsymbol{\rho}) e^{i(\mathbf{k}_1 - \mathbf{k}_2) \cdot \mathbf{R}_n}. \quad (2.5b)$$

The intraband matrix elements may be further approximated as

$$\bar{d}_{cc}(\mathbf{k}_1, \mathbf{k}_2) = \bar{d}_{vv}(\mathbf{k}, \mathbf{k}') \equiv e \frac{\partial}{\partial \mathbf{k}'} \delta(\mathbf{k} - \mathbf{k}'). \quad (2.5c)$$

The optical response of the system requires the calculation of the optical polarization. By quantizing the electromagnetic field and writing down the Heisenberg equation for the field $\hat{E} = [\hat{H} + \hat{H}_F, \hat{E}]$ and $\hat{B} = i[\hat{H} + \hat{H}_F, \hat{B}]$ (\hat{H}_F is the field Hamiltonian), we obtain the Maxwell equation

$$\nabla^2 E(\mathbf{r}, t) - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} E(\mathbf{r}, t) = \frac{4\pi}{c^2} \frac{\partial^2 P(\mathbf{r}, t)}{\partial t^2}, \quad (2.6a)$$

where the polarization operator is given by

$$P(\mathbf{r}, t) \equiv \hat{\psi}^\dagger(\mathbf{r}, t) \mathbf{r} \hat{\psi}(\mathbf{r}, t). \quad (2.6b)$$

Upon the substitution of Eq. (2.2) in Eq. (2.6) and transforming to momentum space, we finally obtain

$$\hat{P}(q) \equiv \int d\mathbf{r} \exp(-iqr) \hat{P}(\mathbf{r}), \quad (2.7a)$$

$$\begin{aligned} \hat{P}(\mathbf{q}) = & \sum_{\mathbf{k}} (\tilde{d}_{cv} \hat{c}_{\mathbf{k}+\mathbf{q}}^\dagger \hat{d}_{\mathbf{k}} + \text{H.c.}) \\ & + \sum_{\mathbf{k}, \mathbf{k}'} [\tilde{d}_{cc}(\mathbf{k}, \mathbf{k}') \hat{c}_{\mathbf{k}+\mathbf{q}}^\dagger \hat{c}_{\mathbf{k}'} \\ & + \tilde{d}_{vv}(\mathbf{k}, \mathbf{k}') \hat{d}_{\mathbf{k}+\mathbf{q}}^\dagger \hat{d}_{\mathbf{k}'}] . \end{aligned} \quad (2.7b)$$

The evaluation of the optical polarization therefore requires the expectation values of the two-body variables:²

$$\hat{Y}_{\mathbf{k}_1 \mathbf{k}_2} \equiv \hat{d}_{\mathbf{k}_1}^\dagger \hat{c}_{\mathbf{k}_2} , \quad (2.8a)$$

$$\hat{C}_{\mathbf{k}_1 \mathbf{k}_2} \equiv \hat{c}_{\mathbf{k}_1}^\dagger \hat{c}_{\mathbf{k}_2} , \quad (2.8b)$$

$$\hat{D}_{\mathbf{k}_1 \mathbf{k}_2} \equiv \hat{d}_{\mathbf{k}_1}^\dagger \hat{d}_{\mathbf{k}_2} . \quad (2.8c)$$

In Appendix C we derive the Heisenberg equations for these operators and close the hierarchy by invoking a factorization of higher-order operators. For simplicity, and similar to what is usually done in the derivation of the SBE,¹ we neglect the intraband dipole couplings. We then obtain the following equations of motion for the two-point electronic correlation function $Y_{\mathbf{k}_1 \mathbf{k}_2}(t) \equiv \langle \hat{Y}_{\mathbf{k}_1 \mathbf{k}_2}(t) \rangle$, $C_{\mathbf{k}_1 \mathbf{k}_2}(t) \equiv \langle \hat{C}_{\mathbf{k}_1 \mathbf{k}_2}(t) \rangle$, and $D_{\mathbf{k}_1 \mathbf{k}_2}(t) \equiv \langle \hat{D}_{\mathbf{k}_1 \mathbf{k}_2}(t) \rangle$:

$$\begin{aligned} \left[i \frac{\partial}{\partial t} - \varepsilon_c(\mathbf{k}_2) - \varepsilon_v(\mathbf{k}_1) \right] Y_{\mathbf{k}_1 \mathbf{k}_2}(t) = & - \sum_{\mathbf{q}} [\tilde{d}_{cv} \mathbf{E}_T(\mathbf{q}, t) \delta_{\mathbf{k}_1, \mathbf{k}_2 + \mathbf{q}} + (\tilde{V}_{\mathbf{q}} + \tilde{V}_{\mathbf{q}}^*) Y_{\mathbf{k}_1 - \mathbf{q}, \mathbf{k}_2 - \mathbf{q}}(t)] \\ & + \sum_{\mathbf{q}} \tilde{d}_{cv} \mathbf{E}_T(\mathbf{q}, t) [C_{\mathbf{q} - \mathbf{k}_1, \mathbf{k}_2}^*(t) + D_{\mathbf{q} - \mathbf{k}_2, \mathbf{k}_1}(t)] \\ & + \sum_{\mathbf{q}, \mathbf{k}} (\tilde{V}_{\mathbf{q}} + \tilde{V}_{\mathbf{q}}^*) [Y_{\mathbf{k}_1 + \mathbf{q}, \mathbf{k} - \mathbf{q}}(t) C_{\mathbf{k}, \mathbf{k}_2}(t) + Y_{\mathbf{k} + \mathbf{q}, \mathbf{k}_2 - \mathbf{q}}(t) D_{\mathbf{k}, \mathbf{k}_1}(t)] , \end{aligned} \quad (2.9a)$$

$$\begin{aligned} \left[i \frac{\partial}{\partial t} - \varepsilon_c(\mathbf{k}_1) + \varepsilon_c(\mathbf{k}_2) \right] C_{\mathbf{k}_1 \mathbf{k}_2}(t) = & - \sum_{\mathbf{q}} [\tilde{d}_{cv} \mathbf{E}_T(\mathbf{q}, t) Y_{\mathbf{q} - \mathbf{k}_2, \mathbf{k}_1 \mathbf{q}}^* - \mathbf{E}_T(\mathbf{q}, t) Y_{\mathbf{q} - \mathbf{k}_1, \mathbf{k}_2}(t)] \\ & + \sum_{\mathbf{k}, \mathbf{q}} (\tilde{V}_{\mathbf{q}} + \tilde{V}_{\mathbf{q}}^*) (Y_{\mathbf{k}, \mathbf{k}_1 + \mathbf{q}}^* Y_{\mathbf{k} + \mathbf{q}, \mathbf{k}_2} - Y_{\mathbf{k}, \mathbf{k}_1}^* Y_{\mathbf{k} + \mathbf{q}, \mathbf{k}_2 - \mathbf{q}}) \\ & + \sum_{\mathbf{k}, \mathbf{q}} \frac{1}{2} \tilde{V}_{\mathbf{q}} (C_{\mathbf{k}_1 + \mathbf{q}, \mathbf{k} + \mathbf{q}} C_{\mathbf{k}, \mathbf{k}_2} - C_{\mathbf{k}, \mathbf{k}_2 - \mathbf{q}} C_{\mathbf{k}_1, \mathbf{k} - \mathbf{q}}) , \end{aligned} \quad (2.9b)$$

$$\begin{aligned} \left[i \frac{\partial}{\partial t} - \varepsilon_v(\mathbf{k}_1) + \varepsilon_v(\mathbf{k}_2) \right] D_{\mathbf{k}_1 \mathbf{k}_2}(t) = & - \sum_{\mathbf{q}} \tilde{d}_{cv} [\mathbf{E}_T(\mathbf{q}, t) Y_{\mathbf{k}_1, \mathbf{k}_2 + \mathbf{q}}^* - \mathbf{E}_T^*(\mathbf{q}, t) Y_{\mathbf{k}_2, \mathbf{k}_1 - \mathbf{q}}] \\ & + \sum_{\mathbf{k}, \mathbf{q}} (\tilde{V}_{\mathbf{q}} + \tilde{V}_{\mathbf{q}}^*) [Y_{\mathbf{k}_1 + \mathbf{q}, \mathbf{k}}^* Y_{\mathbf{k}_2, \mathbf{k} + \mathbf{q}}(t) - Y_{\mathbf{k}, \mathbf{k}_1}^*(t) Y_{\mathbf{k}_2 - \mathbf{q}, \mathbf{k} + \mathbf{q}}(t)] \\ & + \sum_{\mathbf{k}, \mathbf{q}} \frac{1}{2} \tilde{V}_{\mathbf{q}} (D_{\mathbf{k}_1 + \mathbf{q}, \mathbf{k}} D_{\mathbf{k}, \mathbf{k}_2} - D_{\mathbf{k}, \mathbf{k}_2 - \mathbf{q}} D_{\mathbf{k}_1, \mathbf{k} - \mathbf{q}}) . \end{aligned} \quad (2.9c)$$

These equations constitute the main result of this article. They generalize the SBE to include a nonuniform optical excitation and potentially strong dielectric effects. It should be noted that the equations of motion derived directly from the minimal coupling form of the Hamiltonian [Eq. (A1)] are different from the Bloch equations, and do not allow a clear picture of the local-field approximation and its limitations, since they contain the vector potential (A) rather than the transverse electric field.³⁰ This is why we have adopted the dipolar form [Eq. (2.1)].

III. THE LOCAL-FIELD APPROXIMATION FOR HOMOGENEOUS EXCITATION

Equations (2.9) allow the calculation of any four-wave mixing process, including grating experiments that in-

volve a nonuniform excitation. In order to compare with the standard SBE we specialize here to the case of a uniform (homogeneous) excitation. In this case, the electronic correlation functions are independent on the center-of-mass coordinate of the two-point variables. Adopting a notation similar to Ref. 1, we then have

$$Y_{\mathbf{k}_1 \mathbf{k}_2}(t) \cong \delta(\mathbf{k}_1 - \mathbf{k}_2) P_{\mathbf{k}_1}(t) , \quad (3.1a)$$

$$C_{\mathbf{k}_1 \mathbf{k}_2}(t) \cong \delta(\mathbf{k}_1 - \mathbf{k}_2) n_{c\mathbf{k}_1}(t) , \quad (3.1b)$$

$$D_{\mathbf{k}_1 \mathbf{k}_2}(t) \cong \delta(\mathbf{k}_1 - \mathbf{k}_2) n_{v\mathbf{k}_1}(t) , \quad (3.1c)$$

Similar to the standard the SBE, we will assume that the size of the optically active medium is small in comparison with the optical wavelength. Inserting relations (3.1) into Eq. (2.9) we obtain

$$\left[i \frac{\partial}{\partial t} - \varepsilon_c(\mathbf{k}) - \varepsilon_v(\mathbf{k}) \right] P_{\mathbf{k}}(t) = - (n_{c\mathbf{k}} + n_{v\mathbf{k}} - 1) \left[\tilde{d}_{cv} \mathbf{E}_L(t) + \sum_{\mathbf{q}} \tilde{V}_{\mathbf{q}} (P_{\mathbf{k}}^* P_{\mathbf{k} - \mathbf{q}} - P_{\mathbf{k}} P_{\mathbf{k} - \mathbf{q}}^*) \right] , \quad (3.2a)$$

$$i\frac{\partial}{\partial t}n_{ck}(t)=\mathbf{E}_L(t)\tilde{d}_{cv}(P_{\mathbf{k}}-P_{\mathbf{k}}^*)+\sum_{\mathbf{q}}(\tilde{V}_{\mathbf{q}}+\frac{1}{2}\tilde{\tilde{V}}_{\mathbf{q}})(P_{\mathbf{k}}^*P_{\mathbf{k}-\mathbf{q}}-P_{\mathbf{k}}P_{\mathbf{k}-\mathbf{q}}^*). \quad (3.2b)$$

The equation for $n_{vk}(t)$ is identical with right-hand side of Eq. (3.2b), which implies that $n_{ck}(t)=n_{vk}(t)$ within the present approximations. We note that this result does not depend on the assumption of equal effective masses of electrons and holes. The local-field appearing in Eq. (3.2) is given by

$$\mathbf{E}_L(t)\equiv\mathbf{E}_T(r=0,t)+\sum_{\mathbf{q}\neq 0}\tilde{\tilde{V}}_{\mathbf{q}}P_{\mathbf{q}}. \quad (3.3)$$

When the dipole-dipole interaction is neglected (i.e., setting $\tilde{\tilde{V}}_{\mathbf{q}}=0$), we recover the standard SBE (Ref. 1) describing the coupled equations for $P_{\mathbf{k}}, n_{ck}, n_{vk}$ driven by the external transverse field $\mathbf{E}_T(t)$:

$$\left[i\frac{\partial}{\partial t}-\varepsilon_c(\mathbf{k})-\varepsilon_v(\mathbf{k})\right]P_{\mathbf{k}}(t)=-\left(n_{ck}+n_{vk}-1\right)\left[\tilde{d}_{cv}\mathbf{E}_T(t)+\sum_{\mathbf{q}}\tilde{V}_{\mathbf{q}}(P_{\mathbf{k}}^*P_{\mathbf{k}-\mathbf{q}}-P_{\mathbf{k}}P_{\mathbf{k}-\mathbf{q}}^*)\right], \quad (3.4a)$$

$$i\frac{\partial}{\partial t}n_{ck}(t)=\mathbf{E}_T(t)\tilde{d}_{cv}(P_{\mathbf{k}}-P_{\mathbf{k}}^*)+\sum_{\mathbf{q}}\tilde{V}_{\mathbf{q}}(P_{\mathbf{k}}^*P_{\mathbf{k}-\mathbf{q}}-P_{\mathbf{k}}P_{\mathbf{k}-\mathbf{q}}^*). \quad (3.4b)$$

For linear optical response ($n_{ck}=n_{vk}\cong 0$), Eqs. (3.2) can be derived from Eqs. (3.4) by simply replacing the transverse electric field $\mathbf{E}_T(t)$ with the local-field $\mathbf{E}_L(t)$. This is, however, no longer the case for the nonlinear response, since an additional term proportional to $\tilde{\tilde{V}}_{\mathbf{q}}$ appears in Eq. (3.2b) that contributes to the nonlinear response. The fact that the local-field approximation only partially accounts for the dipole-dipole interaction was discussed earlier.³¹ We have shown previously that the third-order nonlinear susceptibility of a semiconductor in the Hartree approximation can be expressed as a sum of contributions resulting from quartic and cubic anharmonicities.³² The additional dipole-dipole contribution in (3.2b) implies that the quartic- and cubic-type terms in the nonlinear susceptibility will scale differently when the dielectric constant is varied.

IV. CONCLUSIONS

In this paper we have derived equations of motion describing two-point, equal-time electronic correlation functions of a two-band semiconductor interacting with a coherent electromagnetic field. Unlike previous treatments of this problem,¹ we have included the dipole-dipole interactions among unit cells and we have not assumed a uniform excitation of the electronic medium. We thus obtained a generalization of the semiconductor Bloch equations (SBE). The two-point electronic dynamics was already considered in Ref. 2 along similar lines; however, the instantaneous Coulomb interaction between electrons was not taken into account in that study.

The equations derived here may provide a starting point for investigations of spectroscopic techniques and limiting cases that are not covered by the ordinary SBE.¹ First, these equations account for the dielectric effect, which may be very strong in semiconductors and could provide an additional contribution to the optical nonlinearity. The external field approximation underlying

the SBE discussed in Ref. 1 was applied to calculations of the optical nonlinearity of thin semiconductor layers, where the local-field correction may be neglected. The other generalization in the present treatment is the consideration of inhomogeneous excitation of the medium, which is necessary for the description of transient grating experiments.²⁸ The present results were obtained within the rigid-lattice approximation (i.e., the electron-phonon interaction was neglected), which is fully justified for off-resonant excitation of the electronic system (phase-grating experiments). When real excitations are present in the medium, the interaction with optical and acoustic phonons cannot be neglected,³³ and Eqs. (2.9) need to be extended to incorporate their effects.

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APPENDIX A: THE DIPOLAR HAMILTONIAN

In this appendix we present the derivation of a dipolar representation of the Hamiltonian of our electronic system interacting with an external classical electromagnetic field. Our main goal is to describe the dipolar approximation when electronic eigenstates are delocalized.

The microscopic minimal coupling Hamiltonian reads⁶

$$H=\int d\mathbf{r}\hat{\psi}^\dagger(\mathbf{r})\left[\frac{1}{2m}(\hat{\mathbf{p}}-e\mathbf{A})^2+\hat{\phi}_c(\mathbf{r})+V_B(\mathbf{r})\right]\hat{\psi}(\mathbf{r}), \quad (A1)$$

where $\mathbf{A}(\mathbf{r},t)$ is the transverse vector potential of the electromagnetic field and we adopt the Coulomb gauge

$\nabla \mathbf{A}(\mathbf{r}, t) = 0$. $V_B(\mathbf{r})$ is the periodic lattice potential, and $\hat{\phi}_c(\mathbf{r})$ is the instantaneous Coulomb potential, e.g., $\hat{\phi}_c(\mathbf{r}) = \int d\mathbf{r}' V_c(\mathbf{r} - \mathbf{r}') \hat{\psi}^\dagger(\mathbf{r}') \hat{\psi}(\mathbf{r}')$. We next perform a gauge transformation:

$$\mathbf{A}(\mathbf{r}, t) \rightarrow \mathbf{A}(\mathbf{r}, t) - \nabla \mathbf{r} \cdot \mathbf{A}(\mathbf{r}, t) \equiv \tilde{\mathbf{A}}(\mathbf{r}, t), \quad (\text{A2a})$$

$$\phi(\mathbf{r}, t) \rightarrow \hat{\phi}_0(\mathbf{r}, t) + \frac{\partial}{\partial t} \mathbf{r} \cdot \mathbf{A}(\mathbf{r}, t) = \tilde{\phi}(\mathbf{r}, t) + \mathbf{r} \cdot \mathbf{E}_T(\mathbf{r}, t), \quad (\text{A2b})$$

which results in

$$H = \int d\mathbf{r} \hat{\psi}^\dagger(\mathbf{r}) \left[\frac{\hat{\mathbf{p}}^2}{2m} + V_B(\mathbf{r}) + \hat{\phi}_c(\mathbf{r}) - \frac{e}{2m} [\hat{\mathbf{p}} \cdot \mathbf{A}(\mathbf{r}, t) + \mathbf{A}(\mathbf{r}, t) \cdot \hat{\mathbf{p}}] + \frac{e^2}{2m} \mathbf{A}^2(\mathbf{r}, t) + \mathbf{r} \cdot \mathbf{E}_T(\mathbf{r}, t) \right] \hat{\psi}(\mathbf{r}). \quad (\text{A3})$$

We rewrite the part of the Hamiltonian that describes the interaction of the electronic system with the transverse and longitudinal electromagnetic field (H_I) as

$$H_I = \sum_n \int_{-R_c/2}^{R_c/2} d\rho \hat{\psi}^\dagger(\mathbf{R}_n + \rho) \left[-\frac{e}{2m} [\hat{\mathbf{p}} \cdot \mathbf{A}(\rho + \mathbf{R}_n, t) + \mathbf{A}(\rho + \mathbf{R}_n, t) \cdot \hat{\mathbf{p}}] + \frac{e^2}{2m} [\tilde{\mathbf{A}}^2(\mathbf{R}_n + \rho, t) + \mathbf{r} \cdot \mathbf{E}_T(\mathbf{R}_n + \rho, t)] \right] \hat{\psi}(\mathbf{R}_n + \rho). \quad (\text{A4})$$

Here, $\mathbf{r} = \mathbf{R}_n + \rho$, where \mathbf{R}_n denotes the center of the n th unit cell and ρ spans the volume of a single unit cell. We next assume that the vector potential does not depend on ρ (i.e., it does not vary within a unit cell). This leads to

$$\tilde{\mathbf{A}}(\rho + \mathbf{R}_n, t) = \mathbf{A}(\mathbf{R}_n, t) - \nabla(\mathbf{R}_n + \rho) \cdot \mathbf{A}(\mathbf{R}_n, t) \cong 0, \quad (\text{A5a})$$

$$\mathbf{E}_T(\mathbf{R}_n + \rho, t) \equiv \frac{\partial}{\partial t} \mathbf{A}(\mathbf{R}_n, t) = \mathbf{E}_T(\mathbf{R}_n, t). \quad (\text{A5b})$$

Inserting relations (A5a) and (A5b) into (A4), we obtain Eq. (2.1).

APPENDIX B: ELECTRON-HOLE REPRESENTATION

In this appendix we discuss the derivation and approximations leading to the form of the Hamiltonian given by Eq. (2.3). We first truncate the multipolar expansion of the Coulomb potential at the dipolar level:

$$V_c(\mathbf{R}_n - \mathbf{R}_m + \rho_1 - \rho_2) \cong \tilde{V}_c(\mathbf{R}_n - \mathbf{R}_m) + (\rho_1 - \rho_2) \cdot \nabla'_{\rho_1} \nabla'_{\rho_2} V_c(\mathbf{R}_n - \mathbf{R}_m + \rho'_1 - \rho'_2) \Big|_{\rho'_1=0, \rho'_2=0}. \quad (\text{B1})$$

Next we split the integration over the spatial variable in H_2 into a sum over the position centers of unit cells \mathbf{R}_n and integrals spanning the volumes of every unit cell:¹

$$H_2 = \sum_{n,m} \int d\rho_1 d\rho_2 \hat{\psi}^\dagger(\mathbf{R}_n + \rho_1) \hat{\psi}^\dagger(\mathbf{R}_m + \rho_2) \hat{\psi}(\mathbf{R}_m + \rho_2) \hat{\psi}(\mathbf{R}_n + \rho_1) V_c(\mathbf{R}_n - \mathbf{R}_m + \rho_1 - \rho_2).$$

We employ the approximation given by Eq. (2.5); we rewrite the part of the Hamiltonian describing the instantaneous Coulomb interaction H_B as

$$H_2 = \frac{1}{2} \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{q}} [\tilde{V}_q(\hat{c}_{\mathbf{k}_1}^\dagger \hat{c}_{\mathbf{k}_2}^\dagger \hat{c}_{\mathbf{k}_2 + \mathbf{q}} \hat{c}_{\mathbf{k}_1 - \mathbf{q}} + \hat{d}_{\mathbf{k}_1} \hat{d}_{\mathbf{k}_2} \hat{d}_{\mathbf{k}_2 + \mathbf{q}}^\dagger \hat{d}_{\mathbf{k}_1 - \mathbf{q}}^\dagger) - (\tilde{V}_q + \tilde{\tilde{V}}_q) \hat{c}_{\mathbf{k}_1}^\dagger \hat{d}_{\mathbf{k}_2} \hat{d}_{\mathbf{k}_2 + \mathbf{q}}^\dagger \hat{c}_{\mathbf{k}_1 - \mathbf{q}}], \quad (\text{B2})$$

where

$$\tilde{V}_q = \sum_n e^{i\mathbf{q} \cdot (\mathbf{R}_n - \mathbf{R}_m)} \tilde{V}_c(\mathbf{R}_n - \mathbf{R}_m), \quad (\text{B3a})$$

$$\tilde{V}_c(\mathbf{R}_n - \mathbf{R}_m) = \frac{e^2}{\epsilon |\mathbf{R}_n - \mathbf{R}_m|}, \quad (\text{B3b})$$

$$\tilde{\tilde{V}}_q = \sum_n e^{i\mathbf{q} \cdot (\mathbf{R}_n - \mathbf{R}_m)} \tilde{\tilde{V}}_c(\mathbf{R}_n - \mathbf{R}_m), \quad (\text{B3c})$$

$$\tilde{\tilde{V}}_c(\mathbf{R}_n - \mathbf{R}_m) = \sum_{i,j} \int d\rho_1 d\rho_2 u_c(\rho_1) \rho_1^i u_v(\rho_1) u_c(\rho_2) \rho_2^j u_v(\rho_2) \frac{\partial^2}{\partial \rho_1^i \partial \rho_2^j} [V_c(\mathbf{R}_n - \mathbf{R}_m + \rho'_1 - \rho'_2)]_{\rho'_1=0, \rho'_2=0}. \quad (\text{B3d})$$

Making these approximations, we obtain

$$\begin{aligned}
H = & \sum_{\mathbf{k}} [E_c(\mathbf{k})\hat{c}_{\mathbf{k}}^\dagger\hat{c}_{\mathbf{k}} - E_v(\mathbf{k})\hat{d}_{\mathbf{k}}\hat{d}_{\mathbf{k}}^\dagger] + \sum_{\mathbf{k},\mathbf{q}} \tilde{d}_{cv} [E_T(\mathbf{q},t)\hat{c}_{\mathbf{k}+\mathbf{q}}\hat{d}_{-\mathbf{k}} + \text{H.c.}] \\
& + \sum_{\mathbf{k},\mathbf{k}'} E_T(\mathbf{k}-\mathbf{k}',t) [\tilde{d}_{cc}(\mathbf{k},\mathbf{k}')\hat{c}_{\mathbf{k}}^\dagger\hat{c}_{\mathbf{k}'} - \tilde{d}_{vv}(\mathbf{k},\mathbf{k}')\hat{d}_{\mathbf{k}}\hat{d}_{\mathbf{k}'}^\dagger] \\
& + \frac{1}{2} \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{q}} [\tilde{V}_q(\hat{c}_{\mathbf{k}_1}^\dagger\hat{c}_{\mathbf{k}_2}^\dagger\hat{c}_{\mathbf{k}_2+\mathbf{q}}\hat{c}_{\mathbf{k}_1-\mathbf{q}} + \hat{d}_{\mathbf{k}_1}\hat{d}_{\mathbf{k}_2}\hat{d}_{\mathbf{k}_2+\mathbf{q}}\hat{d}_{\mathbf{k}_1-\mathbf{q}}) + 2(\tilde{V}_q + \tilde{V}_q^*)\hat{c}_{\mathbf{k}_1}^\dagger\hat{d}_{\mathbf{k}_2}\hat{d}_{\mathbf{k}_2+\mathbf{q}}\hat{c}_{\mathbf{k}_1-\mathbf{q}}] .
\end{aligned} \tag{B4}$$

Finally we change to normal ordering of electrons and holes and neglect the terms in the Hamiltonian obtained from commutators that do not preserve the number of electron-hole pairs as well as a constant term. This results in Eqs. (2.3).

APPENDIX C: TRUNCATION OF THE HIERARCHY

In this appendix we describe the ansatz used to approximate the wave function of the electronic system. We show that this ansatz can be straightforwardly obtained from the exact form of the wave function in the bosonic approximation of the electron-hole pairs. This ansatz allows us to factorize higher-order electronic correlation functions, resulting in a closed hierarchy [Eqs. (2.9)].

When electron hole pairs are treated as bosons, the quantum mechanical state of the electronic medium is a coherent state:

$$|\psi_{\text{coh}}(t)\rangle = \mathcal{N} \exp \left[\sum_{\mathbf{k}_1,\mathbf{k}_2} \varphi_{\mathbf{k}_1\mathbf{k}_2}(t) \hat{Y}_{\mathbf{k}_1\mathbf{k}_2}^\dagger(0) \right] |\Omega\rangle, \tag{C1}$$

where $\varphi_{\mathbf{k}_1\mathbf{k}_2}(t) = \langle \psi_{\text{coh}}(t) | \hat{Y}_{\mathbf{k}_1\mathbf{k}_2} | \psi_{\text{coh}}(t) \rangle$. This can be rearranged in the form

$$|\psi_{\text{coh}}(t)\rangle = \prod_{\mathbf{k}_1,\mathbf{k}_2} \mathcal{N}_{\mathbf{k}_1\mathbf{k}_2} \exp[\varphi_{\mathbf{k}_1\mathbf{k}_2}(t) \hat{Y}_{\mathbf{k}_1\mathbf{k}_2}^\dagger(0)] |\Omega\rangle. \tag{C2}$$

When $\hat{Y}_{\mathbf{k}_1\mathbf{k}_2}$ are no longer approximated as bosonic variables (i.e., they obey Pauli commutations relations),

$$[\hat{Y}_{\mathbf{k}_1\mathbf{k}_2}^\dagger, \hat{Y}_{\mathbf{k}_3\mathbf{k}_4}] = \delta_{\mathbf{k}_1\mathbf{k}_3} \delta_{\mathbf{k}_2\mathbf{k}_4} - \delta_{\mathbf{k}_1\mathbf{k}_4} \delta_{\mathbf{k}_2\mathbf{k}_3} - \delta_{\mathbf{k}_2\mathbf{k}_4} \hat{D}_{\mathbf{k}_1\mathbf{k}_3}, \quad (\hat{Y}_{\mathbf{k}_1\mathbf{k}_2})^n = 0 \quad \text{for } n \geq 2,$$

we can write

$$|\psi_{Q\text{coh}}(t)\rangle = \prod_{\mathbf{k}_1,\mathbf{k}_2} \left[n_{\mathbf{k}_1\mathbf{k}_2}(t) + \frac{\varphi_{\mathbf{k}_1\mathbf{k}_2}(t)}{n_{\mathbf{k}_1\mathbf{k}_2}(t)} \hat{Y}_{\mathbf{k}_1\mathbf{k}_2}^\dagger(0) \right] |\Omega\rangle, \tag{C3}$$

where again $\varphi_{\mathbf{k}_1\mathbf{k}_2}(t) = \langle \psi_{Q\text{coh}}(t) | \hat{Y}_{\mathbf{k}_1\mathbf{k}_2} | \psi_{Q\text{coh}}(t) \rangle$ and $2n_{\mathbf{k}_1\mathbf{k}_2}^2(t) = [1 - 4\varphi_{\mathbf{k}_1\mathbf{k}_2}^2(t)]^{1/2} + 1$. One can verify by inspection that for $\mathbf{k}_2 \neq \mathbf{k}_4$,

$$\langle \psi_{Q\text{coh}}(t) | \hat{C}_{\mathbf{k}_1\mathbf{k}_2} \hat{Y}_{\mathbf{k}_3\mathbf{k}_4} | \psi_{Q\text{coh}}(t) \rangle = \langle \psi_{Q\text{coh}}(t) | \hat{C}_{\mathbf{k}_1\mathbf{k}_2} | \psi_{Q\text{coh}}(t) \rangle \langle \psi_{Q\text{coh}}(t) | \hat{Y}_{\mathbf{k}_3\mathbf{k}_4} | \psi_{Q\text{coh}}(t) \rangle \tag{C4a}$$

and for $\mathbf{k}_2 = \mathbf{k}_4$,

$$\langle \psi_{Q\text{coh}}(t) | \hat{D}_{\mathbf{k}_1\mathbf{k}_2} \hat{Y}_{\mathbf{k}_3\mathbf{k}_4} | \psi_{Q\text{coh}}(t) \rangle = \langle \psi_{Q\text{coh}}(t) | \hat{D}_{\mathbf{k}_1\mathbf{k}_2} | \psi_{Q\text{coh}}(t) \rangle \langle \psi_{Q\text{coh}}(t) | \hat{Y}_{\mathbf{k}_3\mathbf{k}_4} | \psi_{Q\text{coh}}(t) \rangle. \tag{C4b}$$

Finally up to third order in $\varphi_{\mathbf{k}_1\mathbf{k}_2}(t)$ we have

$$\langle \psi_{Q\text{coh}}(t) | \hat{Y}_{\mathbf{k}_1\mathbf{k}_2}^\dagger \hat{Y}_{\mathbf{k}_3\mathbf{k}_4} | \psi_{Q\text{coh}}(t) \rangle \cong \langle \psi_{Q\text{coh}}(t) | \hat{Y}_{\mathbf{k}_1\mathbf{k}_2}^\dagger | \psi_{Q\text{coh}}(t) \rangle \langle \psi_{Q\text{coh}}(t) | \hat{Y}_{\mathbf{k}_3\mathbf{k}_4} | \psi_{Q\text{coh}}(t) \rangle. \tag{C4c}$$

In the following, the ansatz for the wave function given by Eq. (C3) was used, which allowed us to factorize four-point electronic correlation functions [Eqs. (C4)] and derive the nonlinear equations [Eq. (2.9)] from the Heisenberg operator equations [Eq. (2.6)].

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