Coherent Electric-Field Effects in Semiconductors

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We present a microscopic analysis of coherent effects induced by electric fields in photoexcited semiconductors in the presence of the Coulomb interaction. Both the terahertz emission and four-wave-mixing signals arising from Bloch oscillations in a semiconductor superlattice are computed consistently. It is predicted that Bloch oscillations should be observable also for miniband widths on the order of the exciton binding energy.

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Coherent effects induced by static electric fields in photoexcited semiconductors have received considerable interest in recent years. Important examples include the field ionization of excitons [1], virtual photoconductivity [2], electric-field-induced optical rectification at semiconductor surfaces [3,4], the ballistic acceleration of photoinjected electron-hole pairs [5], and Bloch oscillations (BO) in semiconductor superlattices [6,7]. All these effects involve the presence of excitons, which are known to play a dominant role in optical excitations close to the band edge [8]. While the linear optical properties of photoexcited semiconductors in the presence of static electric fields and Coulomb interaction are relatively well understood, theoretical treatments of the nonlinear field-induced coherent effects have either neglected the Coulomb interaction altogether [4,9] or restricted themselves to specific aspects of the excitonic problem [2]. There seems to be a clear need for a comprehensive theoretical approach which would enable one to treat the nonlinear effects in the presence of electric fields under full inclusion of the Coulomb interaction.

In the present Letter, we present a microscopic analysis of coherent effects induced by electric fields in photoexcited semiconductors. Our approach is based on the semiconductor Bloch equations (SBE), which have been used to consistently describe coherent optical phenomena in semiconductors [8]. These equations include the Coulomb effects in the Hartree-Fock approximation, and beyond, depending on the treatment of the carrier relaxation and dephasing processes. The SBE make it possible to evaluate coherent effects to all orders in the optical field. In this Letter we extend the SBE to include the electric field. We demonstrate the usefulness of our approach by calculating the terahertz emission and four-wave-mixing (FWM) signals arising from BO in a semiconductor superlattice. The surprising result of this calculation is that BO should be observable also for miniband widths on the order of the exciton binding energy.

We start from the many-body Hamiltonian for a two-band semiconductor $H = H_{\text{s-p}} + H_{\text{Coul}} + H_{\text{dip}}$, where $H_{\text{s-p}}$ contains single-particle energies $\epsilon_{c,v}(\mathbf{k})$ (c and v corresponding to the conduction and valence bands, respectively), H_{Coul} is the Coulomb many-body Hamiltonian, and H_{dip} is the dipole Hamiltonian given by

$$H_{\text{dip}} = -\sum_{\mathbf{k}} \mu E(t)(a^\dagger_{c\mathbf{k}} a_{v\mathbf{k}} + a^\dagger_{v\mathbf{k}} a_{c\mathbf{k}}) - \sum_{\mathbf{k},\mathbf{k}',\lambda=c,v} \langle \lambda,\mathbf{k}' \mid ie\mathbf{F}(t)\cdot\nabla_{\mathbf{k}} \mid \lambda,\mathbf{k}\rangle a^\dagger_{\lambda\mathbf{k}'} a_{\lambda\mathbf{k}} . \quad (1)$$

The first sum in Eq. (1) describes the coupling of the transverse optical field $E(t)$ to the electron-hole excitations represented by the electron (hole) creation and destruction operators $a^\dagger_{c\mathbf{k}}$ ($a_{v\mathbf{k}}$) and $a_{c\mathbf{k}}$ ($a^\dagger_{v\mathbf{k}}$), respectively, and μ is the optical dipole element. The second sum in Eq. (1) describes the coupling to the longitudinal field $\mathbf{F}(t)$. Following the procedure described in Ref. [8], we derive the dynamic equations for the populations $n_{c,v}(\mathbf{k},t)$ and interband polarizations $P(\mathbf{k},t)$, which now include the electric field:

$$\left[\frac{\partial}{\partial t} - \frac{e}{\hbar}\mathbf{F}(t)\cdot\nabla_{\mathbf{k}} - \frac{i}{\hbar}[e_c(\mathbf{k},t) - e_v(\mathbf{k},t)]\right]P(\mathbf{k},t) = \frac{i}{\hbar}[n_c(\mathbf{k},t) - n_v(\mathbf{k},t)]\,\Omega(\mathbf{k},t) + \left.\frac{\partial P(\mathbf{k},t)}{\partial t}\right|_{\text{coll}},$$

$$\left[\frac{\partial}{\partial t} - \frac{e}{\hbar}\mathbf{F}(t)\cdot\nabla_{\mathbf{k}}\right]n_c(\mathbf{k},t) = \frac{-2}{\hbar}\text{Im}[\Omega(\mathbf{k},t)P^*(\mathbf{k},t)] + \left.\frac{\partial n_c(\mathbf{k},t)}{\partial t}\right|_{\text{coll}}, \quad (2)$$

$$\left[\frac{\partial}{\partial t} - \frac{e}{\hbar}\mathbf{F}(t)\cdot\nabla_{\mathbf{k}}\right]n_v(\mathbf{k},t) = \frac{2}{\hbar}\text{Im}[\Omega(\mathbf{k},t)P^*(\mathbf{k},t)] + \left.\frac{\partial n_v(\mathbf{k},t)}{\partial t}\right|_{\text{coll}}.$$

Here $e_c(\mathbf{k},t) = \epsilon_c(\mathbf{k}) - \sum_{\mathbf{k}'} V(\mathbf{k},\mathbf{k}')n_c(\mathbf{k}',t)$, $e_v(\mathbf{k},t) = \epsilon_v(\mathbf{k}) - \sum_{\mathbf{k}'} V(\mathbf{k},\mathbf{k}')n_v(\mathbf{k}',t)$ are the Coulomb-renormalized energies of electrons and holes, respectively, and $\Omega(\mathbf{k},t) = \mu E(t) + \sum_{\mathbf{k}'} V(\mathbf{k},\mathbf{k}')P(\mathbf{k}',t)$ is the generalized Rabi fre-

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quency; $V(\mathbf{k},\mathbf{k}')$ is the Coulomb interaction. The explicit terms in Eq. (2) denote the results obtained in the time-dependent Hartree-Fock approximation, whereas the subscript coll refers to many-body collision terms beyond the Hartree-Fock approximation, and to other dephasing mechanisms, such as carrier-phonon scattering.

We see in Eq. (2) that the presence of the electric field induces partial derivatives with respect to the carrier momentum. The dynamics induced by the electric field can be treated exactly, for arbitrary field strength, by introducing a moving coordinate frame through $\tilde{t} = t$, $\tilde{\mathbf{k}} = \mathbf{k} + \frac{e}{\hbar}\int^t \mathbf{F}(t')\,dt'$. In this coordinate frame the SBE assume the structure of the field-free case, with the additional condition that the quasimomentum obeys the so-called acceleration theorem $\frac{\partial}{\partial t}\tilde{\mathbf{k}} = \frac{e}{\hbar}\mathbf{F}$. Therefore Eq. (2) also opens up the way to studying the influence of the Coulomb interaction on interesting phenomena like dynamic localization [10,11], which arise when the electric field contains a static and a time-dependent part.

The modified SBE, Eq. (2), include excitonic effects, field ionization of excitons, as well as the full class of coherent many-body effects discussed in the literature [8]. Technically speaking, the equations are integro-differential equations, which can be solved numerically on fast computers. As in the flat-band case, solution of the modified SBE for semiconductors in the presence of an electric field makes it possible to compute coherent effects in ultrafast semiconductor spectroscopy to arbitrary order in the optical field.

One way to calculate nonlinear optical signals from Eq. (2) in the low-excitation limit is to iteratively solve these equations in orders of the optical field, with the initial condition that the semiconductor is in its ground state, i.e., $n_v^{(0)}(\mathbf{k}) = 1$. Iteration for the first two orders yields the following equations of motion for the interband polarization and the populations, respectively:

$$\left[\frac{\partial}{\partial t} - \frac{e}{\hbar}\mathbf{F}(t)\cdot\nabla_{\mathbf{k}} - \frac{i}{h}\epsilon(\mathbf{k})\right]P^{(1)}(\mathbf{k},t) = \frac{-i}{\hbar}\left(\mu E(t) + \sum_{\mathbf{k}'} V(\mathbf{k},\mathbf{k}')P^{(1)}(\mathbf{k}',t)\right) + \left.\frac{\partial P(\mathbf{k},t)}{\partial t}\right|_{\mathrm{coll}}, \tag{3}$$

$$\left[\frac{\partial}{\partial t} - \frac{e}{\hbar}\mathbf{F}(t)\cdot\nabla_{\mathbf{k}}\right]n_{c,v}^{(2)}(\mathbf{k},t) = \frac{\mp 2}{\hbar} \times \mathrm{Im}\left[\left(\mu E(t) + \sum_{\mathbf{k}'} V(\mathbf{k},\mathbf{k}')P^{(1)}(\mathbf{k}',t)\right)P^{(1)*}(\mathbf{k},t)\right] + \left.\frac{\partial n_{c,v}^{(2)}(\mathbf{k},t)}{\partial t}\right|_{\mathrm{coll}}. \tag{4}$$

Here $\epsilon_{cv}(\mathbf{k}) = \epsilon_c(\mathbf{k}) - \epsilon_v(\mathbf{k}) + \sum_{\mathbf{k}'} V(\mathbf{k},\mathbf{k}')$ is the transition energy between conduction and valence band states. Further iteration would lead to equations of motion for higher-order polarizations and populations. An interesting point is that the Coulomb interaction affects interband polarizations and populations in different ways, as can be seen from the structure of Eqs. (3) and (4). In Eq. (3), the Coulomb interaction couples the polarization $P^{(1)}(\mathbf{k},t)$ to all $P^{(1)}(\mathbf{k}',t)$, through the term $\sum_{\mathbf{k}'} V(\mathbf{k},\mathbf{k}')P^{(1)}(\mathbf{k}',t)$. Such couplings exist also for higher-order polarizations. The same Coulomb term in the population equations (4) does not lead to a direct coupling of $n_{c,v}^{(2)}(\mathbf{k},t)$ to $n_{c,v}^{(2)}(\mathbf{k}',t)$, $\mathbf{k} \neq \mathbf{k}'$. The renormalization of the Rabi frequency in Eq. (4) only modifies the population generation. For transient excitation of the semiconductor, this means that after the original generation process the coherent dynamics of the populations is only weakly influenced by the Coulomb interaction; the populations are then determined by the field-induced dynamics. We would hence expect that the signature of the Coulomb interaction should be much stronger in coherent effects associated with interband polarizations than in those determined by the population dynamics.

In the remainder of this Letter we demonstrate the usefulness of our approach by analyzing an example of great current interest, namely the so-called Bloch oscillations. This phenomenon occurs when a crystal electron subjected to a static electric field moves in k space according to the acceleration theorem, and executes a periodic motion in both momentum and real space with a time period T_B, which is inversely proportional to the static electric field. This oscillatory motion translates itself into the frequency domain as a spectrum of equally spaced energy levels, the so-called Wannier-Stark ladder (WSL), with a level spacing of $\frac{h}{T_B}$. Although postulated a long time ago [12], both phenomena have only recently been detected in optical experiments on semiconductor superlattices [6,7,13]. While excitonic effects in linear optical WSL spectra are well understood [14,15], the role of the Coulomb interaction in the nonlinear transient experiments performed to detect the BO has remained unclear. However, one point that seemed to be evident is that the observability of BO requires the existence of a well-defined WSL with an essentially linear dependence of the WSL transition energies on the applied field F. Such WSL have actually been present in the BO measurements in the literature. A different kind of level structure is found in narrow-miniband superlattices, where the miniband width is on the order of the exciton binding energy and the level structure in the linear optical spectra is characterized by an anticrossing behavior [14,16]. We will demonstrate, by solving the modified SBE, that even in this case BO should be observable. This surprising result is a consequence of the different way in which populations and interband polarizations are affected by the Coulomb interaction.

The experimental techniques which have been employed for the detection of BO are transient FWM [6] and terahertz emission spectroscopy [7]. In both cases, the nonlinear signals can be evaluated by solving the SBE iteratively in orders of the optical field. The calculated FWM signal is proportional to the square of the third-order interband polarization $|P^{(3)}(t,\tau)|^2$ in the direction $2\mathbf{k}_2 - \mathbf{k}_1$. Here we denote by $\mathbf{k}_1$ and $\mathbf{k}_2$ the propagation directions of the two laser pulses, $E_j(t)\exp[-i(\mathbf{k}_j \cdot \mathbf{r} - \omega t)]$ ($j = 1,2$), which participate in the generation of the FWM signal; t denotes the real time, and τ the time delay between the two pulses. The terahertz signal is proportional to $\frac{\partial}{\partial t} j^{(2)}(t)$, where the second-order current $j^{(2)}(t)$ is calculated as $j^{(2)}(t) = \frac{e}{\hbar} \int \frac{\partial \epsilon(k)}{\partial k} n^{(2)}(k,t)\, dk$. Additionally, we compute the linear susceptibility $\chi^{(1)}(\omega)$, from which one obtains the linear absorption as $\mathrm{Im}[\chi^{(1)}(\omega)]$.

The model which we employ in our calculation is a one-dimensional tight-binding model of a superlattice with cosine miniband dispersions for the electronic and the heavy-hole minibands [9]. The collision terms in the modified SBE, Eq. (2), are treated as simple exponential relaxations, with times T_1 and T_2 for the populations and the polarizations, respectively [17]. We assume on-site Coulomb interaction, i.e., $V(k,k') = V$ [18]; this form of the Coulomb interaction retains the most important excitonic features, namely a single bound state and an ionization continuum. Our model has the advantage of simplicity and great computational convenience; while it will not be capable of making quantitative statements, it does give qualitatively meaningful results.

We perform calculations with a combined miniband width of $\Delta = 10$ meV; this value is assumed to be field independent as is usually done in tight-binding model calculations for superlattices in an electric field [6,9,19]. We use a Coulomb interaction energy of $V = 9$ meV, which is the binding energy of a single quantum well exciton [20]. The values for the relaxation times employed here are $T_1 = T_2 = 2$ ps. For the calculation of the transient nonlinear signals we use Gaussian laser pulse envelopes $E(t)$ with a duration of 300 fs (FWHM) for the pulse intensity $|E(t)|^2$; the central frequency of the pulses is situated at 2 meV below the excitonic resonance.

Solving the SBE to first order in the optical field, we obtain the linear absorption $\mathrm{Im}[\chi^{(1)}(\omega)]$. Figure 1 shows the maxima of the absorption spectrum in the field range $eFd = 0,...,8$ meV, where F is the applied electric field and d is the superlattice period. The level structure is qualitatively similar to that calculated in an earlier treatment of the excitonic WSL for a narrow-miniband superlattice [14]. A striking feature is the anticrossing between the excitonic resonance and the WSL transitions. This feature is strongly reminescent of the anticrossing behavior observed on $GaAs/Al_{0.3}Ga_{0.7}As$ superlattices with 6 and 11 meV miniband widths [16]. In the following we will investigate the coherent dynamics at one of the anticrossings in Fig. 1. Figure 2 shows the transient FWM signal calculated for an electric field corresponding to $eFd = 3.5$ meV. The signal exhibits a clear modulation, with a time period of about 4 ps, which matches the beat period expected for the energy splitting of 1 meV at the anticrossing. The surprising point is now that we find an altogether different behavior for the terahertz signal, Fig. 3. Here the time period of the modulation is 1.2 ps, in perfect agreement with the BO period of $T_B = 1.2$ ps expected for this value of the electric field. We emphasize that the two signals have been calculated for identical parameters; hence the discrepancy between the modulation periods must have a physical origin.

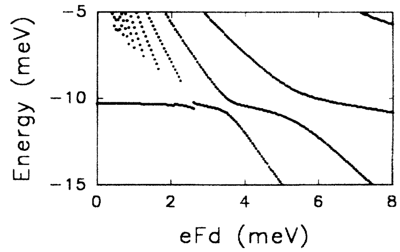

FIG. 1. Fan-chart obtained from the linear absorption $\mathrm{Im}[\chi^{(1)}(\omega)]$ for a superlattice with combined miniband width of $\Delta = 10$ meV. The zero in the energy scale corresponds to a transition to the center of the combined miniband.

The explanation for this behavior lies in the role played by the Coulomb interaction with respect to the different dynamics of populations and interband polarizations as described in the previous sections. The third-order polarization determining the FWM signal is at all times controlled by the Coulomb interaction, and thus fully experiences the presence of the quasibound excitonic resonance. Hence the dynamical evolution of the FWM signal is governed by the energy splitting at the anticrossing between the excitonic resonance and the Wannier-Stark resonance originating from the ionization continuum. The terahertz signal, in contrast, is directly connected to the second-order population, which, in the absence of the driving inhomogeneity in Eq. (4), undergoes a free evolution without being affected by the electron-hole interaction. Thus after completion of the exciting pulse and the decay of the initial first-order polarization, the second-order current, which gives rise to the terahertz signal, evolves as if there was no quasibound excitonic resonance and hence no anticrossing in the WSL chart. In other words, the current is free to perform BO unimpeded by the electron-hole interaction. It is for this reason that the terahertz signal is modulated with the BO frequency.

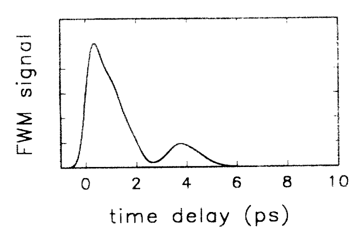

FIG. 2. Calculated transient four-wave-mixing signal as function of time delay for $eFd = 3.5$ meV. We have assumed an inhomogeneous broadening of 2 meV (FWHM).

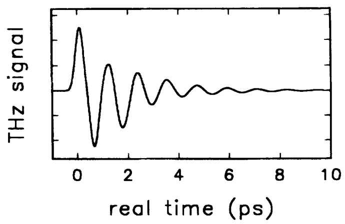

FIG. 3. Calculated terahertz signal as function of real time for the same parameters as in Fig. 2.

It is clear from the aforesaid how one would have to proceed to verify the calculated behavior experimentally. Transient FWM and terahertz emission experiments would have to be performed on a narrow-miniband superlattice with an anticrossing in the linear spectra as described above. Under simultaneous excitation of the resonances involved in the anticrossing, the predicted behavior would show up as a linear dependence of the terahertz-signal modulation frequency on the applied electric field; in contrast, the FWM modulation frequency should go through a minimum at the electric field corresponding to the anticrossing.

In conclusion, we have presented an approach to the theoretical treatment of coherent effects induced by electric fields in photoexcited semiconductors in the presence of the Coulomb interaction. The approach is based on the semiconductor Bloch equations, which we have extended to incorporate the electric field. We have shown that the different way in which populations and interband polarizations are affected by the Coulomb interaction leads to important consequences for the field-induced dynamics of the photoexcited system. We have illustrated this point by calculating the terahertz emission and four-wave-mixing signals arising from Bloch oscillations in a narrow-miniband semiconductor superlattice in the presence of excitonic effects.

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[1] D.F. Blossey and P. Handler, in *Electroabsorption in Modulation Techniques*, edited by R.K. Willardson and A.C. Beer (Academic Press, New York and London, 1972), p. 257.
[2] E. Yablonovitch *et al.*, Phys. Rev. Lett. **63**, 976 (1989).
[3] B.B. Hu, X.-C. Chang, and D.H. Auston, Phys. Rev. Lett. **67**, 2709 (1991).
[4] S.L. Chuang *et al.*, Phys. Rev. Lett. **68**, 102 (1992).
[5] W. Sha *et al.*, Phys. Rev. Lett. **67**, 2553 (1991).
[6] J. Feldmann *et al.*, Phys. Rev. B **46**, 7252 (1992).
[7] C. Waschke *et al.*, Phys. Rev. Lett. **70**, 3319 (1993).
[8] H. Haug and S.W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors* (World Scientific, Singapore, 1993), 2nd ed., and references therein.
[9] G. von Plessen and P. Thomas, Phys. Rev. B **45**, 9185 (1992).
[10] J. Bleuse, G. Bastard, and P. Voisin, Phys. Rev. Lett. **60**, 220 (1988).
[11] D.H. Dunlap and V.M. Kenkre, Phys. Rev. B **34**, 3625 (1986).
[12] M. Holthaus, Phys. Rev. Lett. **69**, 351 (1992).
[13] C. Zener, Proc. R. Soc. London A **145**, 523 (1934); G.H. Wannier, *Elements of Solid State Theory* (Cambridge Univ. Press, Cambridge, England, 1959).
[14] E.E. Mendez, F. Agulló-Rueda, and J.M. Hong, Phys. Rev. Lett **60**, 2426 (1988); P. Voisin *et al.*, Phys. Rev. Lett. **61**, 1639 (1988).
[15] M.M. Dignam and J.E. Sipe, Phys. Rev. Lett. **64**, 1797 (1990); Phys. Rev. B **43**, 4097 (1991).
[16] D.M. Whittaker, Phys. Rev. B **41**, 3238 (1990); R.P. Leavitt, J.L. Bradshaw, and F.J. Towner, Phys. Rev. B **44**, 11 266 (1991).
[17] A.M. Fox *et al.*, Phys. Rev. B **46**, 15 365 (1992).
[18] M. Wegener *et al.*, Phys. Rev. A **42**, 5675 (1990).
[19] S. Schmitt-Rink *et al.*, Opt. Lett. **15**, 60 (1990).
[20] Experimentally, such a system could be realized using, e.g., a $GaAs/Al_{0.3}Ga_{0.7}As$ superlattice with structural parameters close to those given in Ref. [17] for a conduction miniband width of 11 meV. Generally speaking, narrow minibands such as that quoted here can be taylored by using a combination of relatively large barriers and not-too-small well widths.