Intraband Dynamics at the Semiconductor Band Edge: Shortcomings of the Bloch Equation Method

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(Received 11 October 1995)

The validity of the semiconductor Bloch equations (SBE) depends on the approximate decomposition of an intraband correlation function into a product of interband transition densities. We analyze the consequences of this approximation on the intraband dynamics of an optically excited semiconductor. As a special example where the SBE treatment becomes questionable we consider the THz emission of a narrow band superlattice in a static bias field. A comparison of the second order SBE solution with a rigorous second order treatment of this system helps one identify the weak points of the SBE approach and understand the physical background of its failure.

PACS numbers: 71.35.–y, 42.50.Md, 42.65.–k

The semiconductor Bloch equations (SBE) form a cornerstone in the nonlinear optics of semiconductors. The obvious success of these equations in the explanation of numerous effects [1] tends to disguise the fact that the SBE are based on the ill-controlled Hartree-Fock (HF) approximation [2]. The range of validity of the SBE is therefore not clearly defined. In the present Letter we demonstrate that under certain circumstances the approximate character of the SBE may strongly affect the predicted response of the semiconductor already in second order in the driving field. Our example will be the intraband current in a superlattice with narrow minibands excited by a short laser pulse in the presence of a static bias field.

In order to find out whether the signal as derived from the second order SBE solution is influenced by the HF approximation inherent in the SBE we compare it with the complete second order solution of the underlying microscopic model. This is achieved by using the method of *dynamics controlled truncation* (DCT) of the hierarchy of density matrices for optically excited semiconductors. The idea of the DCT method developed in detail in [3,4] relies on the observation that a complete calculation of the nonlinear optical response of a semiconductor to any prescribed order in the driving field can be achieved by considering only a finite set of electronic correlation functions.

To illustrate our point we use the one-dimensional model presented in Ref. [5]. We have chosen this model for two reasons: First, it allows for an easy solution of the relevant set of equations of motion rigorously to second order and, second, it exhibits a qualitatively remarkable feature. According to the SBE treatment the long time behavior of the terahertz emission signal should practically not be affected by the Coulomb interaction between the carriers. In contrast to this, the rigorous second order solution calculated using the DCT method turns out to be noticeably influenced by the Coulomb interaction.

The microscopic basis for the model of Ref. [5] is provided by a Hamiltonian comprising the single particle energies $\epsilon_k^{c,v}$ for conduction and valence bands, respectively, the dipole coupling to the optical field $E(t)$ as well as to a static longitudinal field F and the Coulomb interaction between the particles involved. The $\epsilon_k^{c,v}$ represent nondegenerate minibands with cosine dispersion and combined miniband width Δ. The Coulomb interaction is modeled by a contact potential of strength V.

For the intraband processes the dynamical objects of most interest are the occupation densities for conduction and valence band $C_k := \langle \hat{c}_k^\dagger \hat{c}_k \rangle$ and $D_k := \langle \hat{d}_k^\dagger \hat{d}_k \rangle$, where $\hat{c}_k^\dagger$ ($\hat{d}_k^\dagger$) create electrons (holes) with wave vector k.

Within the DCT approach C_k is derived from the electron hole pair density $N_{\kappa\lambda\mu\nu} := \langle \hat{c}_\kappa^\dagger \hat{d}_{-\lambda}^\dagger \hat{d}_{-\mu} \hat{c}_\nu \rangle$ via the relation

$$C_k = \sum_\lambda N_{k\lambda\lambda k} + \mathcal{O}(E^4). \tag{1}$$

An analogous relation holds for D_k [3,6,7]. It is important to note that (1) does not rely on assumptions about the coherence in the system. In fact, (1) has been derived rigorously in Ref. [4] explicitly taking into account the coupling to a phonon bath in addition to the interparticle interaction, band energies, and spatially dependent band edges. There it has also been discussed in detail why the validity of (1) is not expected to be limited by any of the interaction processes usually considered to be relevant for the description of transient optical experiments near the band edge. In the present paper relation (1) forms the basis of the discussion of intraband dynamics.

Applying the DCT concept to leading order to the one-dimensional model considered in this paper we obtain the following equation of motion for the pair density N, which by construction allows for a calculation of N up to $\mathcal{O}(E^2)$:

$$\{-i\hbar(\partial_t + 1/T_1) + \epsilon^c_\nu + \epsilon^v_{-\mu} - \epsilon^c_\kappa - \epsilon^v_{-\lambda} + ieF(\partial_\kappa + \partial_\lambda + \partial_\mu + \partial_\nu)\} N_{\kappa\lambda\mu\nu}$$
$$+ \sum_q V_q \{N_{\kappa+q,\lambda+q,\mu,\nu} - N_{\kappa,\lambda,\mu-q,\nu-q}\} = M_0 E\, \delta_{\mu\nu} Y^*_{\lambda\kappa} - M^*_0 E^* \delta_{\lambda\kappa} Y_{\mu\nu} + \mathcal{O}(E^4). \quad (2)$$

Here, $V_q = V$ is the contact potential representing the Coulomb interaction in this model, M_0 is the interband dipole matrix element, and $Y_{\mu\nu} := \langle \hat{d}_{-\mu} \hat{c}_\nu \rangle$ is the interband pair transition density.

In order to facilitate the comparison of our calculations with the results of Ref. [5] we not only use the same Hamiltonian with the same parameters as in Ref. [5] but also adopt the method to treat all couplings to bath systems via the introduction of phenomenological dephasing times. It has been pointed out in Ref. [4] that one is not entirely free to choose the dephasing times when it is desired that this phenomenological approach should not be in conflict with microscopic descriptions of the corresponding physical processes. In our case this implies the identification of the dephasing time in the equation for N with the carrier relaxation time T_1, as discussed in Ref. [6]

In the homogeneous situation studied in this Letter the transition density Y can be written as $Y_{\mu\nu} = \delta_{\mu\nu} Y_\mu$. From (2) we therefore find N to be of the form $N_{\kappa\lambda\mu\nu} = \delta_{\kappa\lambda}\,\delta_{\mu\nu} N_{\lambda\mu}$.

Before N can be calculated from (2) to lowest order, one first has to determine Y_k to linear order in the exciting field. The corresponding equation of motion reads

$$\{-i\hbar(\partial_t + 1/T_2) + \epsilon^{cv}_k + ieF\,\partial_k\} Y_k - \sum_q V_q Y_{k-q} = M_0 E, \quad (3)$$

where $\epsilon^{cv}_k := \epsilon^c_k + \epsilon^v_{-k} = \hbar\omega_g - \frac{\Delta}{2}\cos(kd)$.

Inverting (3) in terms of the relevant Green function we obtain

$$Y_k(t) = M_0 \int_{-\infty}^t dt'\, G_k(t - t', T_2)\, E(t'). \quad (4)$$

An explicit representation for the Coulomb Green function G_k is easily constructed along the lines described in Ref. [5]. Using the Coulomb Green function $G_{kk'}$ as being the response to the source $\delta(t)\,\delta_{kk'}$, the lowest order solution for N according to Eq. (2) reads

$$N_{k\lambda}(t) = i\hbar \int_{t'} \sum_{k'\lambda'} G^*_{kk'}(t - t', 2T_1)\, G_{\lambda\lambda'}(t - t', 2T_1) \times [M_0 E(t') Y^*_{k'}(t') - M^*_0 E^*(t') Y_{\lambda'}(t')]. \quad (5)$$

With the help of (1), (4), (5), and the identity $\sum_\lambda G_{k\lambda}(\tau, T_0)\, G_\lambda(T, T_0) = \frac{i}{\hbar}\theta(\tau)\,\theta(T)\, G_k(T + \tau, T_0)$, which can easily be derived using the eigenfunction representation for the Green functions G_k and $G_{k\lambda}$, we obtain for the electronic occupation density

$$C_k(t) = |M_0|^2\, 2 \int_{-\infty}^t dt' \times \mathrm{Re}\Big[G^*_k(t - t', T_2)\, E^*(t')\, e^{-(1/T_1 - 2/T_2)(t-t')} \times \int_{-\infty}^{t'} dt''\, G_k(t - t'', T_2)\, E(t'') \Big] + \mathcal{O}(E^4). \quad (6)$$

From C_k the THz signal then is found as in Ref. [5].

Figure 1 shows THz signals that have been calculated for the same parameters as in Ref. [5]. Along with the DCT result derived from (6), we have plotted curves according to different approximate treatments. The curve marked "$V = 0$" corresponds to the case of strictly vanishing Coulomb interaction, while the result of the semiconductor Bloch equations is labeled SBE. As predicted in previous papers [5,8,9] the SBE signal becomes more and more similar to the Coulomb free case when the optical pulse as well as the interband polarization have decayed. On the other hand, the qualitative differences to the DCT result are obvious. Another limiting case of interest is the so-called *coherent limit* [6]. It has been shown in Ref. [6] that, provided the dephasing times fulfill the relation $2T_1 = T_2$, the occupation density is given by $C_k = |Y_k|^2 + \mathcal{O}(E^4)$. This strict relation between the dephasing times is, however, in most cases not a realistic assumption. Having calculated the complete second order intraband response for our model offers the opportunity to test the validity of an approximation scheme that keeps the same simple functional relation between C_k and Y_k as in the coherent limit. The different time scales of interband and intraband dephasing processes are corrected by hand according to the ansatz

$$C_k(t) \xrightarrow{\mathrm{TCCL}} e^{-(1/T_1 - 2/T_2)t} |Y_k(t)|^2. \quad (7)$$

From (6) it follows that this *time scale corrected coherent limit* (TCCL) becomes exact to lowest order not only in the coherent limit, but also in the limit of an ultrashort optical pulse. As can be seen from Fig. 1 for a 300 fs full width half maximum (FWHM) pulse the TCCL curve still follows very closely the DCT result.

We will now try to localize the discrepancy between the SBE and the rigorous DCT solution. The equation of motion for C_k according to the SBE is given by

$$\{-i\hbar(\partial_t + 1/T_1) + ieF\partial_k\} C_k = 2\,i\,\mathrm{Im}\Big\{\Big[E(t)M_0 + \sum_q V_{k-q} Y_q(t) \Big] Y^*_k(t)\Big\}, \quad (8)$$

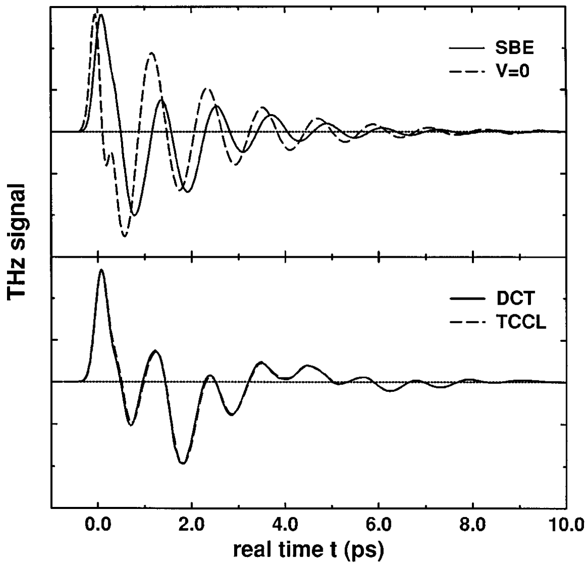

FIG. 1. Calculated THz signals. Upper part: solid line, SBE treatment; dashed line, Coulomb-free limit. Lower part: solid line, DCT result according to Eq. (6); dashed line, TCCL approximation; cf. (7). The parameters were $\Delta = 10$ meV, $V = 9$ meV, $T_1 = T_2 = 2$ ps, $Fed = 3.5$ meV. The pulses were chosen to be of Gaussian shape, with 0.3 ps FWHM for the intensity and a central frequency 2 meV below the exciton resonance.

which differs from the corresponding DCT equation only by the last term on the right hand side in the way that the four point function N has been replaced by Y^*Y. Such a factorization, however, is only correct in the coherent limit. Consequently, in this limit the SBE solution coincides with the DCT response. Discussing the implications of the above factorization, we shall concentrate on two aspects: the frequency contents and the time scales involved. First we analyze the frequency contents.

From relation (1) it follows that the second order intraband signal exhibits excitonic features on any time scale because the propagation of N according to Eq. (2) is generated by the difference of two excitonic Hamiltonians and the relevant sources are excitonic too. Hence the DCT approach predicts no single particle signatures in the THz signal. This result is not specific of the simple one-dimensional model treated in this Letter. It holds whenever the fundamental relation (1) is true and is especially independent of the form of the interaction potential. Using, e.g., a screened potential would affect the excitons and their spectra, but not the fact that no single particle energies enter the THz signal. What is specific of the present model is that the intraband frequencies are given as differences of the resonances in the linear spectrum. In more general situations this feature will be lost. So, e.g., the coupling to a phonon reservoir will renormalize the relevant frequencies and thus destroy the above mentioned parallelism between interband and intraband dynamics [4]. Thus in a thorough analysis of the frequency contents of an observed signal one has to be aware of the possibility that dissipative interactions might influence the result. Such effects are neglected in our present model because we have used phenomenological dephasing times. Furthermore, when the frequency contents of intraband observables is compared with other signals dominated by interband processes like four wave mixing (FWM) measurements, there are additional sources for differences, as the FWM signal is influenced by renormalizations due to two pair scattering states as well as to contributions from bound biexcitons already to leading order in the optical field, while corresponding terms would enter the THz signal only via higher order contributions.

Let us now discuss the frequency contents of the HF approximate solution derived from Eq. (8). For short pulse excitations the source proportional to $E(t)$ is practically a δ function, while the Y^*Y source behaves like a step function modulated by oscillations with differences of the excitonic transition frequencies. Under these conditions the $E(t)$ source will excite oscillations with frequencies determined by the homogeneous part and these are single particle frequencies not affected by the Coulomb interaction. The Y^*Y source will excite a superposition of forced oscillations with differences of excitonic transition frequencies and free oscillations with single particle frequencies such as to guarantee continuity at the onset of the pulse. And here comes the great temptation: Looking at the SBE solution one expects to see the free particle signature in the THz signal when the interband source Y^*Y has decayed. As we have already shown, this feature is not reproduced by the complete second order solution. Remembering that the only difference between the DCT and the SBE approach in the present context amounts to a replacement of the four point density N in the equation for C_k by its factorized counterpart Y^*Y, we must conclude that retaining the unfactorized source proportional to N results in a cancellation of the apparent single particle oscillations. Obviously there exists a delicate balance which is disturbed by the HF approximation. This problem, by the way, is common to all approaches that approximately replace N by something else in the equation for the intraband density C_k. Examples are the introduction of Boltzmann type scattering terms or the Kadanoff-Baym ansatz. Whether or not the replacement of the N source leads to noticeable errors depends on the context. Especially when many resonant states or even a continuum of states are involved, the manipulation of the source might be harmless. Another case where the factorization of N is justified has already been mentioned, namely, a system near the coherent limit. Here is the point where the aspect of time scales comes in.

According to the fundamental relation (1), which holds even in the presence of a dissipative reservoir, the N source will never decay faster than C_k. Thus also the long time behavior will be characterized by forced excitonic oscillations in contrast to the SBE approach where of all contributions only those appear to be long living, which in the consequent DCT treatment will be canceled. The pitfall in the SBE treatment is the replacement of a long living density like source N) by a short living transition like quantity Y^*Y).

From these considerations we conclude that deviations of the SBE approach from the rigorous solution should be most pronounced (1) when the excitation is selective to states strongly affected by excitonic effects and (2) when the system is far from the coherent limit. This is confirmed by the results of our calculations shown in Fig. 2. Compared with Fig. 1 we have taken spectrally narrower pulses 0.8 ps FWHM), centered on the exciton line (cf. the fan chart in Ref. [5]). Furthermore, we have chosen values for the dephasing times far from the coherent limit. For these excitation conditions the DCT solution almost exclusively shows oscillations with a frequency of about 4 ps corresponding to the energy splitting for the lowest levels in the anticrossing region. In contrast, the SBE result in the long-time regime approaches the Coulomb free limit exhibiting weak oscillations with the single particle Bloch frequencies. Also displayed is the TCCL result. As expected the quantitative deviations from the DCT solution are now noticeable because of the longer pulse length, but the qualitative behavior is still quite similar to the DCT curve. The TCCL treatment, therefore, provides a simple approximation scheme that can be expected to yield reasonable results in many cases, especially for short pulse excitation, provided a model with constant dephasing times is appropriate.

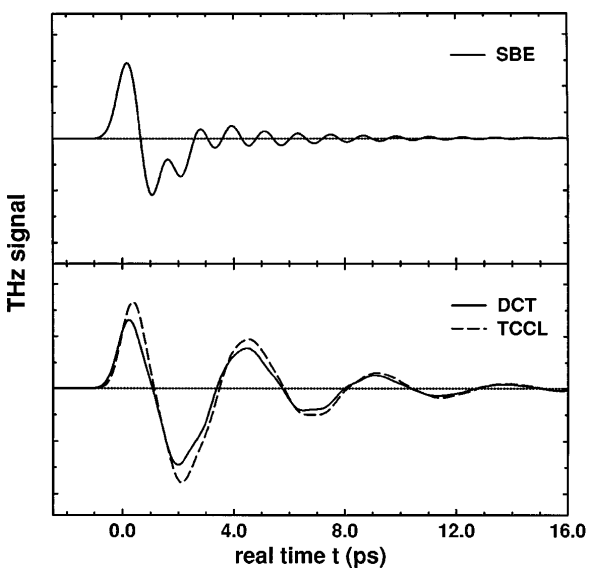

FIG. 2. Calculated THz signals as in Fig. 1. The parameters are the same as in Fig. 1 except for $T_1 = 4$ ps, a pulsewidth of 0.8 ps FWHM for the intensity and the central frequency in resonance with the exciton.

Let us finally have a look at the experimental situation and ask to what extent our analysis is confirmed. So far most observations of THz signals have been reported to be interpretable in terms of excitonic frequencies within experimental errors [9–11]. Nevertheless, there are also reports that frequency components different from the ones occurring in interband observables are present [8,12]. But whatever the physical origin of these new oscillation frequencies will finally turn out to be, an explanation based on the SBE must not be trusted even in cases where predictions extracted from the SBE solution do not explicitly contradict the experimental findings, because with respect to this particular aspect the SBE are in qualitative disagreement with the underlying microscopic model they aim to approximate.

We thank K. Victor for useful discussions and P. Haring Bolivar and H. G. Roskos for clarifying the experimental situation.

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