Nonperturbative scaling behavior of the coherent semiconductor Bloch equations in the low-density regime

Th. Östreich and A. Knorr*
Institut für Theoretische Physik, Universität Göttingen, Bunsenstraße 9, D-37073 Göttingen, Federal Republic of Germany
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The set of phenomenological model equations which we proposed to determine the scaling behavior of the low-density Rabi oscillations [Phys. Rev. B **48**, 17811 (1993)] is justified from the coherent part of the semiconductor Bloch equations in lowest order in the light-matter coupling. In contrast to similar short-time studies, it is shown that for resonant excitation of an excitonic state the Rabi oscillations can be described by a Ginzburg-Landau type equation which includes only the exciton-exciton interaction of the resonantly excited state but no state-filling terms or contributions from detuned excitonic or continuum resonances. Explicit calculations are performed for the bulk two-band jellium model and for a one-dimensional two-band Hubbard model.

I. INTRODUCTION

In a previous publication[1] we proposed the following set of phenomenological equations to determine the oscillation frequency and amplitude of low-density Rabi oscillations in a semiconductor instead of solving the full semiconductor Bloch equations (SBE):

$$\begin{aligned}\dot{S}_1 &= -\left[\delta + J_1(1+S_3)\right] S_2 \ , \\ \dot{S}_2 &= \left[\delta + J_2(1+S_3)\right] S_1 + J_3 S_3 \ , \\ \dot{S}_3 &= \Omega S_2 \ .\end{aligned} \tag{1}$$

The equations (1) contain only macroscopic variables, i.e., the polarization components $S_{1(2)}$ and the inversion density S_3 of the semiconductor, the Rabi frequency Ω, the detuning from the $1s$ exciton resonance δ, and a phenomenological parameter vector $\mathbf{J} = (J_1, J_2, J_3)$. In the limit of low electron-hole pair density, we estimated the parameters of the $\mathbf{J}$ model to be

$$\begin{aligned} J_1 = J_2 \equiv J_\perp \ , \\ J_3 = \Omega. \end{aligned} \tag{2}$$

Here, $J_\perp$ remains as a fitting parameter. In this regime of low electron-hole pair density, the inversion S_3 can be approximated with $P = \frac{1}{2}(S_1 - iS_2)$ by $S_3 \approx -1 + 2|P|^2$ and we find an equation for the complex polarization P alone:

$$\dot{P} = -i(\delta + 2J_\perp |P|^2)P + i\Omega/2. \tag{3}$$

We compared this model with the solution of the coherent part of the semiconductor Bloch equations (SBE) for zero detuning $\delta = 0$ and showed that in the limit of small Rabi frequency Ω the frequency and the amplitude of the semiconductor Rabi oscillations are well described by fitting the dynamics with the parameter $\mathbf{J}$. Only for a simplified model system with large Coulomb on-site interaction was the parameter $J_\perp$ determined by the atomic transition frequency Δ_x, where the semiconductor ground state and the excitonic ground state are degenerate.

In this paper we show how to calculate the phenomenological $\mathbf{J}$ parameter in lowest order [cf. Eq. (2)] microscopically by applying an expansion of the coherent SBE for low-excitation processes (see, for instance, Ref. 2). In addition to this we justify the neglect of nonresonant contributions for the description of Rabi oscillations quantitatively. The applied approach was already used for the investigation of the *short-time* behavior of the SBE in the context of photon echo studies,[3] the optical Stark effect,[4] and for traveling wave solutions in self-induced transparency of excitonic systems.[5–7] Starting with the full coherent SBE for arbitrary semiconductor structures,[8–11] the derivation of a Ginzburg-Landau-type equation for the polarization with effective control parameters, which are completely determined microscopically, was demonstrated.

For the short-time behavior a perturbation theory for this equation with respect to powers of the Rabi frequency may be applied. This approach breaks down for the long-time behavior[3] and especially on a time scale where Rabi oscillations of the total electron-hole density occur. In contrast to the short-time expansion, where the leading contribution of the resulting polarization scales linearly with Rabi frequency, a *different* expansion with respect to the Rabi frequency for the *long time* behavior must be applied. In addition to this, we show explicitly that for resonant excitation of the $1s$ exciton nonresonant contributions vanish in the limit of vanishing ratio of Rabi frequency and exciton binding frequency Ω/ω_x. Furthermore, we will show that for resonant excitations this expansion yields a scaling of the total electron-hole density amplitude with the Rabi frequency which indicates that space filling effects play a minor role in comparison to the exciton-exciton interaction. This result explains our previous results on nonlinear Rabi oscillations in this regime and justifies the use of the proposed simplified dynamical system.[1]

II. EFFECTIVE DYNAMICAL EQUATIONS

In the coherent regime, i.e., neglecting any dephasing mechanism, the dynamics of the expectation values of the microscopic polarization functions $P_{\mathbf{q}} = \langle c^{\dagger}_{1,\mathbf{q}} c_{2,\mathbf{q}} \rangle$ and the population inversion $w_{\mathbf{q}} = \langle n_{2,\mathbf{q}} - n_{1,\mathbf{q}} \rangle$ is given in the time-dependent Hartree-Fock approximation by[8–11]

$$\frac{\partial P_{\mathbf{q}}(t)}{\partial t} = -i \left(\Delta_{\mathbf{q}} - \omega_p - \frac{1}{\hbar} \sum_{\mathbf{q}'} \tilde{U}_{\mathbf{q}-\mathbf{q}'} w_{\mathbf{q}'}(t) \right) P_{\mathbf{q}}(t) - i \left(\frac{\Omega}{2} + \frac{1}{\hbar} \sum_{\mathbf{q}'} \tilde{U}_{\mathbf{q}-\mathbf{q}'} P_{\mathbf{q}'}(t) \right) w_{\mathbf{q}}(t) \, , \tag{4}$$

$$\frac{\partial w_{\mathbf{q}}(t)}{\partial t} = 2i P^{*}_{\mathbf{q}}(t) \left(\frac{\Omega}{2} + \frac{1}{\hbar} \sum_{\mathbf{q}'} \tilde{U}_{\mathbf{q}-\mathbf{q}'} P_{\mathbf{q}'}(t) \right) + \text{c.c.} \tag{5}$$

The Rabi frequency $\Omega = \frac{2\mu E_0(t)}{\hbar}$ is determined by the interband dipole matrix element μ and the slowly varying amplitude of the external electrical field $\mathcal{E}(t) = E_0(t) e^{-i\omega_p t}$. The electron-hole pair dispersion relation $\Delta_{\mathbf{q}}$, as well as the Fourier transform of the Coulomb interaction $\tilde{U}_{\mathbf{q}} = \frac{1}{V} \int U(\mathbf{x}) e^{-i\mathbf{q}\cdot\mathbf{x}} d^3x$, depends on the specific realization of the semiconductor model.

Now the main steps of the reduction of the SBE, which can be found, for instance, in Ref. 3, are briefly reviewed. In the weak nonlinear regime, the density of the optically excited electron-hole pairs is assumed to be small or equivalently the inversion $w_{\mathbf{q}}(t)$ is almost -1. It was shown[2] that using the constants of motion of the SBE (probability conservation), it is possible to expand the inversion in terms of the polarization with the condition $|P_{\mathbf{q}}|^2 \ll 1$:

$$\begin{aligned} w_{\mathbf{q}} &= -\sqrt{1 - 4|P_{\mathbf{q}}|^2} \\ &= -1 + 2|P_{\mathbf{q}}|^2 + 2|P_{\mathbf{q}}|^4 + O(|P_{\mathbf{q}}|^6). \end{aligned} \tag{6}$$

In this regime, Eq. (5) may be dropped and the SBE reduce to a set of equations for the polarization alone. With the help of the eigenstates of the Wannier operator

$$(H_x)_{\mathbf{q},\mathbf{q}'} = \left(E_G + \hbar(\Delta_{\mathbf{q}} - \Delta_{\mathbf{q}=0}) + \sum_{\mathbf{q}''} \tilde{U}_{\mathbf{q}-\mathbf{q}''} \right) \times \delta_{\mathbf{q},\mathbf{q}'} - \tilde{U}_{\mathbf{q}-\mathbf{q}'} \tag{7}$$

defined by $\sum_{\mathbf{q}'} (H_x)_{\mathbf{q},\mathbf{q}'} \alpha_{\mathbf{q}',n} = E_x^{(n)} \alpha_{\mathbf{q},n}$, we may expand the microscopic polarization functions $P_{\mathbf{q}}$ as

$$P_{\mathbf{q}}(t) = \sum_n \alpha_{\mathbf{q},n} P_n(t). \tag{8}$$

Inserting this ansatz in Eq. (4) and multiplying both sides with $\sum_{\mathbf{q}} \alpha^{*}_{\mathbf{q},\lambda}$ one finds with $\sum_{\mathbf{q}} \alpha^{*}_{\mathbf{q},\lambda} \alpha_{\mathbf{q},\lambda'} = \delta_{\lambda,\lambda'}$ a set of equations for the coefficients P_λ. Besides the low-density expansion $|P_k|^2 \ll 1$ another essential approximation, which underlies the reduction of the full set of SBE, is the restriction to an excitonic bound state in the case of near resonant excitation, i.e., if the laser central frequency is tuned at a selected exciton resonance $\omega_p \equiv E_x^{(n)}/\hbar$ and the Rabi frequency Ω and the spectral pulse width are much less than the energy spacing of the neighboring levels. It is well known that for resonant excitation with $\delta_0 = E_x^{(0)}/\hbar - \omega_p \approx 0$ the polarization distribution is shown to exhibit the characteristic behavior of the exciton ground state wave function $P_{\mathbf{q}} \sim \alpha_{\mathbf{q},0} P_0(t)$ during temporal evolution under weak optical excitation.[12] At the critical point $\delta_0 = 0$, the fundamental resonance is separated by the order of the exciton binding energy $\hbar\omega_x$ from the higher excitonic resonances. We will demonstrate how this fundamental frequency enters the scaling of the reduced equation, and that in the long-time behavior the nonresonant contributions really vanish not only due to the decreasing density of states but due to a different scaling with respect to the Rabi frequency. With the approximation $\alpha_{\mathbf{q},n} \approx \alpha_{\mathbf{q},n} \delta_{n,\lambda}$ Eq.(4) is simplified to

$$\begin{aligned} \frac{\partial P_\lambda}{\partial t}(t) = &-i(E_x^{(\lambda)}/\hbar - \omega_p) P_\lambda(t) + \frac{2i}{\hbar} \sum_{\mathbf{q},\mathbf{q}'} \tilde{U}_{\mathbf{q}-\mathbf{q}'} \left(|\alpha_{\mathbf{q},\lambda}|^2 |\alpha_{\mathbf{q}',\lambda}|^2 - |\alpha_{\mathbf{q},\lambda}|^2 \alpha^{*}_{\mathbf{q},\lambda} \alpha_{\mathbf{q}',\lambda} \right) |P_\lambda|^2 P_\lambda \\ &+ i \frac{\Omega}{2} \left(\sum_{\mathbf{q}} \alpha^{*}_{\mathbf{q},\lambda} - 2 \sum_{\mathbf{q}} |\alpha_{\mathbf{q},\lambda}|^2 \alpha^{*}_{\mathbf{q},\lambda} |P_\lambda|^2 - 2 \sum_{\mathbf{q}} |\alpha_{\mathbf{q},\lambda}|^4 \alpha^{*}_{\mathbf{q},\lambda} |P_\lambda|^4 \right) \\ &+ \frac{2i}{\hbar} \sum_{\mathbf{q},\mathbf{q}'} \tilde{U}_{\mathbf{q}-\mathbf{q}'} \left(|\alpha_{\mathbf{q},\lambda}|^2 |\alpha_{\mathbf{q}',\lambda}|^4 - |\alpha_{\mathbf{q},\lambda}|^4 \alpha^{*}_{\mathbf{q},\lambda} \alpha_{\mathbf{q}',\lambda} \right) |P_\lambda|^4 P_\lambda. \end{aligned} \tag{9}$$

Furthermore, in the limit $\Omega/\omega_x \ll 1$, which we discuss here the phase space filling term $\sim \Omega |P|^4$ in Eq. (9) can already be neglected in contrast to the contribution $\sim \tilde{U}_{\mathbf{q}} |P|^4$. Introducing a rescaled polarization variable

$$P(t) = \frac{1}{\sum_{\mathbf{q}} \alpha^{*}_{\mathbf{q},\lambda}} P_\lambda(t) \, , \tag{10}$$

we finally get the following expression, using $\delta_\lambda = E_x^{(\lambda)}/\hbar - \omega_p$:

$$\dot{P} = -i\delta_\lambda P + i \frac{\Omega}{2} \left(1 - \beta_{2,\lambda} |P|^2 \right) - i \left(\beta_{1,\lambda} |P|^2 + \beta_{3,\lambda} |P|^4 \right) P \, , \tag{11}$$

with the parameters $\alpha_\lambda = \sum_{\mathbf{q}} \alpha^*_{\mathbf{q},\lambda}$ and

$$\beta_{1,\lambda} = \frac{2|\alpha_\lambda|^2}{\hbar} \sum_{\mathbf{q},\mathbf{q}'} \tilde{U}_{\mathbf{q}-\mathbf{q}'} \times \left(|\alpha_{\mathbf{q},\lambda}|^2 \alpha^*_{\mathbf{q},\lambda}\alpha_{\mathbf{q}',\lambda} - |\alpha_{\mathbf{q},\lambda}|^2|\alpha_{\mathbf{q}',\lambda}|^2\right) , \quad (12)$$

$$\beta_{2,\lambda} = 2\alpha_\lambda \sum_{\mathbf{q}} |\alpha_{\mathbf{q},\lambda}|^2 \alpha_{\mathbf{q},\lambda}, \quad (13)$$

$$\beta_{3,\lambda} = \frac{2|\alpha_\lambda|^4}{\hbar} \sum_{\mathbf{q},\mathbf{q}'} \tilde{U}_{\mathbf{q}-\mathbf{q}'} \times \left(|\alpha_{\mathbf{q},\lambda}|^4 \alpha^*_{\mathbf{q},\lambda}\alpha_{\mathbf{q}',\lambda} - |\alpha_{\mathbf{q},\lambda}|^4|\alpha_{\mathbf{q}',\lambda}|^2\right) . \quad (14)$$

Equation (11) is a Ginzburg-Landau-type equation[13] for a macroscopic variable of the system. Similar equations have been derived in the context of self-induced transparency of Wannier excitons[3,5,6,14,15] and Frenkel excitons.[16] The control parameters in Eqs. (12)–(14) are microscopically well defined and can be calculated even analytically in some cases where the exciton ground state wave function is known.

Now the $J_\perp$ parameter may be calculated by comparing Eq. (3) with Eqs. (11) and (12) for different semiconductor systems rigorously. In principle, higher order corrections to the full parameter vector $\mathbf{J}$ in Eq. (1) can be derived in a similar fashion, but the calculation becomes somewhat involved. It is therefore more appealing to introduce $\mathbf{J}$ *a posteriori* as a phenomenological parameter vector, to be determined, for instance, experimentally.

In our previous paper[1] we discussed two general cases with respect to the fundamental $1s$-exciton resonance ($\beta_i = \beta_{i,0}$).

(i) $\delta_0 \neq 0$. Slightly off-resonant but still fulfilling $\delta_0 \ll \omega_x$ the polarization is of order $P = O(\Omega)$. This was checked numerically and can be estimated by inspecting the stationary solution of Eq. (9). In this case

$$\beta_3|P|^4P = O(|\Omega|^5) , \quad (15)$$

and it is sufficient to retain only β_1 and β_2 to include the first nonlinear corrections because both contributions are of the same order $O(|\Omega|^3)$. However, the leading contribution is the linear term, which introduces the detuning frequency δ_0 as the fundamental frequency in the system. In this case the semiconductor behaves like an atomic system[1] and the linear perturbation theory in the Rabi frequency remains valid.

(ii) $\delta_0 \equiv 0$. The more interesting case is exact resonance, where we observed Rabi oscillations nonlinear in amplitude and frequency with respect to the Rabi frequency. Here, the polarization shows a different scaling behavior and nonlinear corrections become important. From the stationary solution of Eq. (11), one finds for a small Rabi frequency

$$|P|^3 = \frac{\Omega}{2\beta_1}\left(\frac{1-\beta_2|P|^2}{1+\frac{\beta_3}{\beta_1}|P|^2}\right) = \frac{\Omega}{2\beta_1} + O(|\Omega|^{5/3}). \quad (16)$$

The scaling behavior shows clearly that the contribution $\sim \beta_2$ is of the same order as the term $\sim \beta_3$. The leading term of course is the dynamical detuning $\sim \beta_1$ which remains in the limit $\Omega/\beta_1 \ll 1$. At low densities, the neglect of state-filling terms at exact resonance is therefore well justified. This explains the correct scaling behavior of the nonlinear electron-hole pair oscillations in optically excited semiconductors as shown previously[1] with an effective nonlinear dynamical system.

The contribution of any nonresonant term which scales like Ω/ω_x can be neglected in comparison to Eq. (16). This reveals a different quality in contrast to the photon echo studies for the short-time behavior, where the nonresonant terms are neglected only due to their smaller density of states, whereas these contribution really vanish in the discussed limit of Rabi oscillations for very small Rabi frequency. The discussion of the two different cases shows that the exciton-exciton interaction of the resonant excited state contributes at higher order in Ω to the signal amplitude than the state-filling and the off-resonant excited states. We present some results for the microscopic parameters of two semiconductor systems, where the integrals Eqs. (12)–(14) have either been calculated in a different context[3,17,18] or can be calculated analytically as in the case of the one-dimensional Hubbard model.

III. TWO-BAND JELLIUM MODEL

The two-band jellium model is commonly used in the study of optical properties of semiconductor bulk or quantum well systems. The model contains the two parameters of the exciton binding frequency ω_x and the Bohr radius $a_0 = \frac{e^2}{2\hbar\omega_x}$ of the ground state wave function of the Wannier operator Eq. (7). For the bulk system the Fourier transform of the Coulomb potential and the hydrogen wave function for the $1s$ exciton are given by

$$\alpha_{\mathbf{q},0} = \frac{1}{\sqrt{V}} \frac{8\sqrt{\pi a_0^3}}{[1+(\mathbf{q}a_0)^2]^2}, \qquad \tilde{U}_{\mathbf{q}} = \frac{4\pi e^2}{V\mathbf{q}^2}. \quad (17)$$

With this information, the microscopic parameters Eqs. (12) and (13) can be calculated analytically for $\alpha_0 = \sqrt{V/(\pi a_0^3)}$:

$$\beta_1 = \tfrac{26}{3}\omega_x, \qquad \beta_2 = 7 . \quad (18)$$

The additional parameter β_3 can also be calculated analytically and we find $\beta_3 = \frac{2^{19}}{1470}\omega_x$. This term together with β_2 are the first $1s$ corrections in the reduced dynamical system Eq. (11). It has to be clearly pointed out that corrections from higher excitonic states as well as the continuum states contribute at this order concerning the expansion parameter Ω/ω_x.

IV. TWO-BAND HUBBARD MODEL

In order to cover a one-dimensional exactly solvable semiconductor model as well, we give the results for a two-band tight-binding Hubbard model with on-site interaction energy U_0 and band parameter B. In this case, the system has only one bound state with wave function[19]

$$\alpha_{\mathbf{q},0} = \frac{1}{\sqrt{V}} \frac{U_0}{\sqrt{U_0^2 + B^2} - B\cos\mathbf{q}} \left[1 + \left(\frac{B}{U_0}\right)^2\right]^{-1/4},$$

$$\tilde{U}_{\mathbf{q}} = \frac{U_0}{V}, \tag{19}$$

and exciton binding energy $\hbar\omega_x = \sqrt{U_0^2 + B^2} - B$. The control parameters are given by

$$\alpha_0 = \sqrt{V}\left[1 + \left(\frac{B}{U_0}\right)^2\right]^{-1/4}, \tag{20}$$

$$\beta_1 = \frac{U_0}{\hbar}\left(\frac{B}{U_0}\right)^2\left[1 + \left(\frac{B}{U_0}\right)^2\right]^{-3/2}, \tag{21}$$

$$\beta_2 = 3 - \left[1 + \left(\frac{B}{U_0}\right)^2\right]^{-1}. \tag{22}$$

In the limit of large on-site interaction $B \ll U_0$, the exciton-exciton interaction parameter β_1 is asymptotically given by $\beta_1 \approx \frac{B^2}{\hbar U_0}$. This result can be related to the critical atomic energy separation $\hbar\Delta_x \to \frac{B^2}{2U_0}$, where the intrinsic ground state of the semiconductor, i.e., the completly filled valence band state, and the exciton ground state are degenerate. In Ref. 1, the value of the parameter $2J_\perp \equiv \beta_1$ was estimated to be Δ_x in the strong coupling limit, as can now be proved analytically, too.

In conclusion, we have justified the usage of the simplified **J** model from the reduction of the SBE to a simplified dynamical system, which is estimated to be valid in the limit of low electron-hole density for near resonant excitation of the fundamental exciton resonance. At exact resonance, i.e., $\hbar\omega_p = E_x^{(0)}$, only one control parameter β_1 is sufficient to yield asymptotically exact results in the limit $\Omega/\beta_1 \ll 1$ and confirms previous results on nonlinear Rabi oscillations in semiconductors.[1,20] In addition, we have shown that not only phase space filling contributions $\sim \beta_2$ but also higher exciton-exciton corrections $\sim \beta_3$ are of the same order of magnitude at exact resonance, which has not been noted in earlier results.[5]

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* Present address: Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität, Renthof 5, D-35032 Marburg, Federal Republic of Germany.

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