Asymptotic analytic solution for Rabi oscillations in a system of weakly excited excitons

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An asymptotic solution for light-induced exciton density oscillations in a semiconductor is calculated from the semiconductor Bloch equations in the low density limit (weak optical excitation). The analytic approximation is compared with a numerical solution of the full equations. It is shown that even for low area fields ($\ll 2\pi$) density oscillations are caused by Coulomb exchange effects coupling the generated excitons and that the oscillation frequency and amplitude depend nonlinearly on the Rabi frequency of the incident field.

It is well known that coherent pumping of atomic systems with an optical field $Ee^{-i\omega t}$ (carrier frequency ω) yields temporal oscillations in the electron density (Rabi oscillations) if the exciting optical pulses E exceed a certain area. The area Θ of a square pulse is defined by the product of the pulse duration t_L and Rabi frequency Ω of the light field:

$$\Theta = \Omega t_L\,, \quad \Omega = \frac{Ed}{\hbar}, \tag{1}$$

where d is the dipole moment of the optical transition. An analytic solution for the electron density n of the corresponding atomic (two level) Bloch equations for the case of resonant excitation is given by[1]

$$n = \sin^2\left(\frac{\Omega t}{2}\right). \tag{2}$$

From this equation it can be seen that a pulse area larger than 2π yields Rabi oscillations in the temporal development of the electron density. A similar type of such oscillations is observed if a semiconductor is resonantly excited at the exciton resonance.[2,3] Even if the exciton resonance shows a two-level-like behavior in the linear spectrum, there is a main difference concerning the physical interactions of optically excited atomic systems and semiconductor materials: the optically generated excitons in a semiconductor may interact with each other by Coulomb exchange forces. This is not possible for the electrons in atomic systems well separated from each other. This additional interaction of the generated excitons due to exchange interaction in the semiconductor yields additional effects in the light-matter interaction.[2–4] Therefore the semiconductor has to be described by a type of Bloch equations different from atomic systems. The coherent part of the semiconductor Bloch equations (SBE) contains, in comparison to the atomic Bloch equations, an effective source term and the so-called band gap renormalization.[2] These terms make an analytical treatment of the time dependent SBE very difficult and numerical methods have to be applied to calculate Rabi oscillations.[3] Analytic solutions for the SBE exist for stationary situations and nonstationary situations with vanishing exchange contributions (for a review see Ref. 4) or for times after the optical excitation.[5] The numerical solutions[3] for the nonstationary case including exchange effects show significant differences between the semiconductor and atomic Rabi oscillations. In the atomic case Rabi oscillations are harmonic oscillations and the oscillation frequency for resonant excitation is determined by the value of the Rabi-frequency Ω exclusively; compare Eq. (2). In contrast to this, for resonant excitation of the exciton level of a semiconductor a second fundamental frequency comes into play: the exciton binding frequency ω_{exc} which determines the strength of the exchange effects.

In this paper we derive an asymptotic analytic solution for Rabi oscillations of an exchange coupled system of excitons at low density. In our approximation scheme we take into account only the $1s$ exciton and neglect the continuum states as well as higher excitons. It will turn out that the low density limit is connected to the situation where the Rabi frequency of the light Ω Eq. (1) is much smaller than the exciton binding frequency ω_{exc}. After the derivation of the time dependent low density solution from the semiconductor Bloch equations we compare the obtained analytic results with numerical solutions of the full semiconductor Bloch equations to clarify the influence of higher exciton and continuum states.

Instead of writing down the full SBE (Ref. 2) we only present the Bloch equations for the leading exciton resonance as derived in Refs. 2 and 6. These equations are a good approximation for low excitation phenomena, which were already shown in the studies of the semiconductor photon echo.[6] In this context the interaction of a semiconductor with light is described by an equation for the dimensionless polarization function P of the $1s$ exciton only. The equation may be written in the form

$$\frac{\partial P}{\partial t} = i\frac{\Omega}{2} - i\beta_1|P|^2P - i\beta_2|P|^2\frac{\Omega}{2}. \tag{3}$$

Here β_1 is proportional to the exciton binding frequency ω_{exc} (Ref. 7) and β_2 is a dimensionless constant. For the usual two-band model[2,8] one obtains $\beta_1/\omega_{\text{exc}} = 26/3$ and $\beta_2 = 7$. The total polarization is obtained by multiplying the polarization function with the Sommerfeld-factor of $2/(\pi {a_0}^3)$, where a_0 is the excitonic Bohr radius and the factor of 2 counts for the spin summation. Note that in the wave number representation of the SBE, which includes in contrast to Eq. (3) high density effects, the validity of Eq. (3) is restricted to a small modulus of the polarization P_k of a one-particle state with wave number k: $|P_k|^2 \ll 1$.[2,6,10] In our approximation the dimensionless polarization function P for the $1s$ exciton is connected to P_k by the following expression:

$$P_k \approx \frac{\psi_k^{1s}}{\sqrt{\pi {a_0}^3}} P, \tag{4}$$

where ψ_k^{1s} is the wave function of the first exciton in k-space.

The polarization function P in Eq. (3) is driven by the optical field Ω, the exciton-exciton interaction (Coulomb exchange contributions) is described by the second term and the exciton-photon interaction (state filling) yields the third term in Eq. (3). The exciton-photon interaction is small compared to the exciton-exciton interaction in the limit of a small ratio of Rabi frequency and exciton binding frequency. The consistency of this assumption is shown later by inserting the calculated solution Eq. (11) into Eq. (3). However a rough estimate of the polarization amplitude may be obtained by the stationary solution of Eq. (3):

$$|P|^2 = \left(\frac{\Omega}{2\beta_1}\right)^{\frac{2}{3}} \left\{1 + O\left[\beta_2 \left(\frac{\Omega}{2\beta_1}\right)^{\frac{2}{3}}\right]\right\}. \tag{5}$$

Hence we neglect the exciton-photon interaction in the asymptotic limit of a small ratio of Rabi frequency and exciton binding frequency. From Eq. (3) we find a set of equations for the real and imaginary parts of the polarization $P = u + iv$ and for the quantity $n = |P|^2$, which is proportional to the total electron-hole density:

$$\frac{\partial u}{\partial t} = \beta_1 n v\,, \quad \frac{\partial v}{\partial t} = \frac{\Omega}{2} - \beta_1 n u\,, \quad \frac{\partial n}{\partial t} = \Omega v. \tag{6}$$

By inserting Eq. (6c) in Eq. (6a) and by differentiating Eq. (6c) we derive, under the assumption that Ω is given by a square pulse, an equation of motion for n, which is equivalent to a quartic oscillator model:

$$\frac{\partial^2 n}{\partial t^2} = \frac{\Omega^2}{2}\left(1 - n^3 \frac{\beta_1^2}{\Omega^2}\right). \tag{7}$$

Integrating this equation in the usual way and writing $n = (2\Omega/\beta_1)^{2/3}\tilde{n}$ we obtain a first order differential equation for $\tilde{n}$ in the dimensionless time variable $x = \Omega t(\beta_1/2\Omega)^{\frac{1}{3}}$:

$$\frac{\partial \tilde{n}}{\partial x} = \left(\tilde{n} - \tilde{n}^4\right)^{\frac{1}{2}}. \tag{8}$$

Equation (8) can be integrated and yields an expression for the exciton density n:

$$n = \left(\frac{2}{\gamma}\right)^{\frac{2}{3}} \frac{1 - cn(3^{\frac{1}{4}}x, k)}{\sqrt{3} + 1 + (\sqrt{3} - 1)cn(3^{\frac{1}{4}}x, k)}\,, \quad \gamma = \frac{\beta_1}{\Omega}. \tag{9}$$

The cn-functions are the Jacobian functions with modulus $k = \sin(\pi/12)$.[9] The cn-function is a periodic function and can be written in terms of trigonometric functions for small $k^2 = 0.067$:

$$cn(x, k) = \cos(x) + O(k^2). \tag{10}$$

Hence the expression for n takes the final form

$$n = \left(\frac{2}{\gamma}\right)^{\frac{2}{3}} \frac{1}{1 + \sqrt{3}\cot^2\left(\frac{3^{\frac{1}{4}}x}{2}\right)}. \tag{11}$$

With this expression for n a rather simple approximation for the total electron-hole density $2n/\pi {a_0}^3$ of the full SBE for the low density limit is found, which takes only the $1s$ exciton state into account. Note that our procedure does not imply a perturbation theory in time; a solution valid for arbitrary times is derived.

To clarify the validity of our approach for the full SBE and to investigate the influence of the neglected higher exciton and continuum states we have solved the coherent part of the SBE (Ref. 2) numerically for different parameters γ. The values of the parameter γ for which Eq. (11) yields a good approximation to the full numerical solutions of the SBE is determined by the condition $|P_k|^2 \ll 1$ [as mentioned following Eq. (3) (Ref. 10)]. Using the wave function of the $1s$ exciton $\psi_k^{1s} = 8\sqrt{\pi {a_0}^3}/[(ka_0)^2 + 1]^2$ we estimate the maximum value of $|P_k|^2$ from Eqs. (4) and (11) as follows:

$$\begin{aligned} |P_k|^2 &\approx \left(\psi_k^{1s}\right)^2 P^2 \frac{1}{\pi {a_0}^3} \\ &= \frac{64n}{[(ka_0)^2 + 1]^4} \\ &\le 64\left(\frac{2}{\gamma}\right)^{\frac{2}{3}}. \end{aligned} \tag{12}$$

We obtain the γ parameter range for the validity of Eq. (11) from $|P_k|^2 \ll 1$:

$$\gamma \gg 2(64)^{\frac{3}{2}} \approx 10^3. \tag{13}$$

Therefore from $\gamma = 10^4$ to higher values a reasonable agreement of the full numerical solution and the analytic solution Eq. (11) is expected. The set of material parameters used in our numerical solution corresponds to bulk CdSe ($\omega_{\text{exc}} = 0.0243\,\text{fs}^{-1}$). The numerical solutions for different values of the parameter $\gamma = 0.5, 2, 10^4$ (dashed lines) of the full SBE for the density $\tilde{n}$ as function of the normalized time coordinate x and one oscillation period of the analytical solution (solid line) are depicted in Fig. 1. From a comparison one can find that as expected

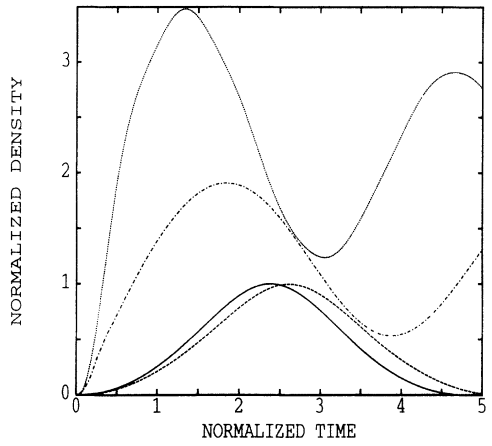

FIG. 1. Numerical solution of the coherent part of the SBE for the density $\tilde{n}$ versus normalized time x for different $\gamma = 0.5, 2, 10^4$ (dashed lines in order of decreasing amplitude) in comparison with the analytic solution Eq. (10) (solid line).

the agreement of the analytic and the numerical solution for increasing γ becomes better. A slight difference in the oscillation frequency is seen to arise for the largest $\gamma = 10^4$ value, which we attribute to the influence of the neglected states which are taken fully into account by the numerical solution but not by our analytical approach. We conclude that the asymptotic solution is very useful to investigate the limit $\gamma^{-1} \to 0$ of the coherent part of the full SBE, especially the amplitude and frequency of the Rabi oscillations.

In the following we show a significant difference of semiconductor Rabi oscillations in comparison to atomic ones: for large γ the product of the time for one complete Rabi-flop and the field amplitude Ω (the area) can be much smaller than 2π but Rabi-flopping is still observed in contrast to atomic systems where an optical pulse with an area smaller than 2π would not induce a complete flop in the electron density. To illustrate this we discuss one characteristic example for bulk CdSe ($\omega_{\mathrm{exc}} = 0.0243\,\mathrm{fs}^{-1}$). For the γ-parameter of $\gamma = 10^3$ and $\Omega = 10^{-3}$ fs^{-1} the period time of the Rabi oscillation is about 10^3 fs yielding a pulse area of $\Theta \approx 1 < 2\pi$ which produces one complete Rabi-flop. For larger values of γ the pulse area which induces on semiconductor Rabi-flop can be much smaller than unity. This can be easily seen by extracting the amplitude A and the frequency ω_{nl} of the nonlinear density oscillations from Eq. (11):

$$A = \left(\frac{2}{\gamma}\right)^{\frac{2}{3}}, \qquad \omega_{\mathrm{nl}} = 3^{\frac{1}{4}} \left(\frac{\gamma}{2}\right)^{\frac{1}{3}} \Omega. \tag{14}$$

From the scaling of the frequency it can be recognized that the semiconductor Rabi oscillations like the atomic Rabi oscillations depend on the Rabi frequency of the light pulse but the oscillation frequency is renormalized by a factor depending on γ which in our approximation is large compared to unity. This implies that complete Rabi-flops may occur for pulses with areas much less than 2π.

Furthermore we can see from the scaling of the amplitude that the exciton-photon interaction is of minor importance compared to the exciton-exciton interaction in Eq. (3):

$$|P|^2 \sim \left(\frac{1}{\gamma}\right)^{\frac{2}{3}}, \quad \gamma P|P|^2 \sim 1. \tag{15}$$

We confirm the approximation made after Eq. (3), that in the limit $\gamma^{-1} \to 0$ the exciton-photon interaction may be neglected. For $\gamma^{-1} \to 0$, the nonlinear density oscillations of Eq. (11) are a unique signature of the coherent exciton-exciton interaction.

It is interesting to note that already for $\gamma = 2$ the analytical solution yields a good approximation for the value of the nonlinear frequency (Fig. 1). For this reason the well known doubling of the Rabi-flop number compared to the atomic case known for 2π and 3π pulses as observed in Refs. 3 and 10 for resonant excitation of the $1s$ exciton may be explained by Eq. (14). For this purpose the ratio of the nonlinear frequency ω_{nl} and the Rabi-frequency Ω of the light field which determines the oscillation frequency in the atomic case is calculated:

$$\frac{\omega_{\mathrm{nl}}}{\Omega} = 3^{\frac{1}{4}} \left(\frac{13\omega_{\mathrm{exc}}}{3\Omega}\right)^{\frac{1}{3}} \approx 2.15 \left(\frac{\omega_{\mathrm{exc}}}{\Omega}\right)^{\frac{1}{3}}. \tag{16}$$

For the situation investigated numerically in Refs. 3 and 10, the Rabi frequency of the light and the exciton binding frequency are approximately in the same order of magnitude, i.e., $\Omega \approx \omega_{\mathrm{exc}}$, a doubling of the number of Rabi-flops occurs due to the factor of approximately 2 in Eq. (16).

In conclusion we have obtained analytic results for resonant low density Rabi-flopping in an exciton system for arbitrary times. The Rabi oscillations are caused by light pulses with areas much less than 2π. This is not possible in atomic systems. The reason was shown to be the interaction of excitons due to the Coulomb exchange effects. The temporal development of the observed Rabi oscillations can be described very well by a simple analytic formula which takes into account the exciton-exciton interaction.

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[7] Equation (3) arises from the expansion of the distribution function f_k with respect to the polarization functions P_k in k space, where k is the wave number: $f_k = |P_k|^2 + |P_k|^4 + O(|P_k|^6)$. We note that for the long time behavior discussed here the second term of the expansion yields higher contributions in Eq. (3) which are on the same order as the term proportional to β_2. These additional terms and the β_2 term are neglected in the following.

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