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2001 J. Phys.: Condens. Matter 13 275

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Excitonic insulator through coherent pulse excitation?

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Received 12 October 2000

Abstract

The excitonic insulator was predicted numerous times by theory, but the experimental observation is conspicuous by absence. We demonstrate that, even under ideal circumstances, an excitonic insulator cannot be created by means of coherent optical excitation. If dephasing is neglected, a non-stationary excitonic insulator may arise. If a dephasing is taken into account, a hypothetical excitonic insulator would disappear within the characteristic dephasing time. Moreover, we find that dephasing prevents the buildup of the excitonic insulator from the beginning.

1. Introduction

The theory of superconductivity by Bardeen, Cooper, and Schrieffer (BCS theory) was a breakthrough in theoretical solid-state physics, and was successful in explaining basic statistical properties of superconductors [1]. A key result of the BCS theory is the observation that an arbitrarily small attraction between electrons of opposite spin leads to an energy gap and to pairing states.

The above theoretical concepts were introduced to semiconductor physics in the sixties [2]. Formal analogy to the BCS theory led to the prediction of an ‘excitonic phase’ in semimetals and small-gap semiconductors. Since excitons, the analog of Cooper pairs, are electrically neutral, this state is often referred to as an excitonic insulator. Intuitively it is plausible that, if the forbidden gap E_g is smaller than the exciton binding energy E_B , rearrangement of the dispersions should take place, otherwise the excited state would be below the ground state.

Shortly afterwards, an excitonic phase was also predicted for highly excited semiconductors with a finite stationary density of electrons and holes [3–6]. The theory goes alongside the theory for small-gap semiconductors, except that the electron and hole dispersions are renormalized by the chemical potentials μ_e and μ_h , and the main results can be adopted if the forbidden gap E_g is replaced by $E_g - \mu_e - \mu_h$.

The realization of short laser pulses, the improvement of the sample quality, and the development of low-dimensional semiconductors fostered the interest in those coherent states of electrons and light in semiconductors that were previously reported only in atomic and molecular systems. In 1986, the optical Stark effect was measured on GaAs/(Ga,Al)As quantum wells by Mysyrowicz *et al* [7], and was explained by Schmitt-Rink and co-workers on the basis of the semiconductor Bloch equations [8]. Very recently, another coherent effect, a

tiny non-resonant contribution below the pump frequency, ‘hyper-Raman gain,’ was observed in bulk GaAs [9]. The theory, also based upon the semiconductor Bloch equations, perfectly reproduces the experimental results [10].

The semiconductor Bloch equations in the absence of an electromagnetic field give rise to a gap equation—the same as in the theory of the excitonic insulator. This time, the forbidden gap has to be replaced by $E_g - \hbar\omega_p$, where ω_p is the frequency of the laser. Thus the photon energy acts like a chemical potential, a fact which has been elaborated in detail by Comte and Mahler [11, 12]. This theoretical result may suggest that an excitonic insulator can be created by excitation of a semiconductor with coherent light.

After this progress, it looks like the experimental observation of the excitonic insulator is at hand. During the last ten years, the number of publications on subjects such as excitonic insulators, Bose condensation, and superfluidity of excitons has dramatically increased [13–18]. However, much confusion results from the fact that authors do not always clearly distinguish between those concepts. Part of the reason is that some early works stated that excitonic insulator and Bose condensation of excitons would be the same [3, 6]. A recent monograph is devoted to studying the similarities and differences of the various condensation phenomena [19].

Let us focus on the excitonic insulator, which has been studied theoretically in recent years by Glutsch, Östreich, Chu, Zhu, Ferraz, Shi, Fernández-Rossier, and others [13–16]. The optical absorption has been calculated several times [13–15]. For resonant excitation, the optical spectrum is characterized by a strong maximum above and a gain region below the effective chemical potential. The results are qualitatively the same for three-, two- and one-dimensional semiconductors.

The correspondence between superconductor and excitonic insulators was emphasized by many authors. Yet this formal analogy is seductive. Let us briefly point out some important differences.

- (a) The superconductor is the *absolute* ground state and the time scale for a conductor to reach the superconducting state is not crucial at all. In contrast, the excitonic insulator is an *intermediate* state and the formation of an excitonic insulator competes with relaxation processes which attempt to bring the system into the ground state.
- (b) In the theory of superconductivity the approximate Hamiltonian \hat{H}_{BCS} does not commute with the number operator $\hat{N} = \hat{N}_\uparrow + \hat{N}_\downarrow$, but the exact Hamiltonian is particle-conserving. The success of the grand-canonical formalism, despite $[\hat{H}_{\text{BCS}}, \hat{N}] \neq 0$, is likely due to ‘cancellation of errors.’ The situation is completely different for the excitonic insulator: the total number of electrons and holes is not conserved, neither exactly, nor in the Hartree–Fock approximation. Therefore, the common practice of introducing a grand-canonical Hamiltonian for highly excited semiconductors is not backed up by the underlying physics.
- (c) In a superconductor, the electron-phonon scattering is incorporated into the effective electron–electron interaction. Ideally, the BCS state is not subjected to dissipation. In contrast, the excitonic insulator is solely an effect of Coulomb attraction. Hence the excitonic insulator still faces dissipation due to electron–phonon scattering, and the order of magnitude is the same as for independent electrons and holes. It is also worthwhile to note that subtracting the chemical potentials from the dispersions in the case of a highly-excited semiconductor is equivalent to a rotating frame [5]. The supposed stationary state is then characterized by an oscillating polarization and, consequently, by radiative decay.

For the above reasons, the process of how the semiconductor evolves into the state of an excitonic insulator is crucial. The situation is different for highly-excited and coherently-driven semiconductors. In a highly-excited semiconductor the initial situation is characterized

by Fermi seas of electrons and holes. The history, how the electrons and holes were created in the first place, is irrelevant. It does not make a difference, for example, whether the carriers are induced by light or injected by electric currents. In a coherently driven excitonic insulator, the coherent state is induced by a laser pulse and the electron-photon system remains coherent all the time until the excitonic insulator is established. There is however a number of limitations. Interactions beyond mean-field theory, such as electron-phonon and exciton-exciton interaction lead to dephasing which inevitably destroys the polarization. For acoustic phonons, this effect is still present in the low-density limit and at liquid helium temperature. In order to comply with the gap equation, which assumes a sharp photon energy, the pump pulse has to be spectrally narrow. On the other hand, a long pulse will increase the risk of dephasing and relaxation.

To the best of our knowledge, there are only two publications that explicitly deal with the time evolution of a coherently driven excitonic insulator. Glutsch and Zimmermann presented time-dependent solutions for the density matrix in the absence of incoherent processes [13]. For resonant excitation, they observed a transition into an excitonic insulator, but the time is much longer than any realistic coherence times. The correctness of this result has been questioned by Östreich and Schönhammer on the basis of energy conservation [15]. At the same time, they proposed another class of solutions, the ‘non-stationary excitonic insulator’ which is characterized by a frequency different from the pump frequency. It was demonstrated for a simplified model that a non-stationary excitonic insulator can be achieved by coherent optical excitation.

In this paper we calculate the time evolution of the density matrix for a large variety of detunings, pulse lengths, and amplitudes. The calculations are done (i) neglecting any incoherent processes, thus assuming the best case, and (ii) with phenomenological dephasing. For no set of parameters an excitonic insulator is obtained. We also show that the lifetime of a hypothetical excitonic insulator is no longer than the dephasing time. The paper is organized as follows: in section 2 we introduce the basic notations, equations, and the numerical algorithm; the results of the calculation are shown in section 3; and a summary and conclusions are given in section 4.

2. Basic equations

The theoretical analysis is based upon density matrix theory. The density matrix in Bloch representation is defined as

$$N_{jj'}(\mathbf{k}) = \langle \hat{a}_{j'\mathbf{k}}^\dagger \hat{a}_{j\mathbf{k}} \rangle \quad (1)$$

where $\hat{a}_{j\mathbf{k}}^\dagger$ ($\hat{a}_{j\mathbf{k}}$) is the creation (annihilation) operator of a Bloch electron with band index j and wave-vector \mathbf{k} .

We consider a direct two-band semiconductor. The conduction (c) and valence (v) band dispersions are assumed to be parabolic, i.e. $E_{c,v}(\mathbf{k}) = E_{c,v}(0) \pm \hbar^2 k^2 / 2m_{e,h}$ with effective electron and hole masses $m_{e,h} > 0$. The transition from valence to conduction band shall be optically allowed with a dipole matrix element μ . The pump field is assumed to have the form $E_p(t) e^{-i\omega_p t} + \text{c.c.}$, where ω_p is the pump frequency and $E_p(t)$ is a slowly varying amplitude. Furthermore, we assume zero temperature so that, in the case of no optical excitation, the valence band is completely occupied and the conduction band is completely empty. The equation of the density matrix in the rotating frame is given by the semiconductor Bloch equations [8]

$$i\hbar \frac{\partial}{\partial t} N(\mathbf{k}, t) = [H(\mathbf{k}, t), N(\mathbf{k}, t)] + i\hbar \frac{\partial}{\partial t} N(\mathbf{k}, t) \Big|_{\text{corr}} \quad (2)$$

$$H(\mathbf{k}, t) = \begin{pmatrix} E_c(\mathbf{k}) - \frac{1}{2}\hbar\omega_p & -\mu E_p(t) \\ -\mu^* E_p^*(t) & E_v(\mathbf{k}) + \frac{1}{2}\hbar\omega_p \end{pmatrix} - \int \frac{d^3\mathbf{k}'}{(2\pi)^3} V(\mathbf{k}-\mathbf{k}') [N(\mathbf{k}', t) - N^{(0)}(\mathbf{k}')]$$

$$\lim_{t \rightarrow -\infty} N(\mathbf{k}, t) = N^{(0)}(\mathbf{k}) = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}.$$

The statically screened Coulomb potential is $V(\mathbf{k}) = e^2/\varepsilon_0\varepsilon k^2$, where ε is the background dielectric constant of the semiconductor.

First, we study the properties of equation (3) in the absence of correlation, i.e. when $\partial N/\partial t|_{\text{corr}} = 0$. Then the trace and the determinant of $N(\mathbf{k}, t)$ are conserved for each \mathbf{k} . Therefore, we may write the density matrix in the form:

$$N(\mathbf{k}, t) = \begin{pmatrix} n(\mathbf{k}, t) & \psi(\mathbf{k}, t) \\ \psi^*(\mathbf{k}, t) & 1 - n(\mathbf{k}, t) \end{pmatrix} \quad (3)$$

where n is referred to the occupation of the conduction band and ψ is called the microscopic polarization. These functions are related to each other by $n(\mathbf{k}, t) [1 - n(\mathbf{k}, t)] = |\psi(\mathbf{k}, t)|^2$ for each \mathbf{k} . Important macroscopic quantities, which can be calculated from the elements of the density matrix, are the electron density

$$\rho(t) = \int \frac{d^3\mathbf{k}}{(2\pi)^3} n(\mathbf{k}, t) \quad (4)$$

which is equal to the hole density or the density of electron–hole pairs, and the macroscopic polarization

$$P(t) = \mu^* \int \frac{d^3\mathbf{k}}{(2\pi)^3} \psi(\mathbf{k}, t). \quad (5)$$

Strictly speaking, since we have introduced the rotating frame, P is the slowly varying amplitude of the macroscopic polarization.

Before we turn to the time-dependent solutions of equation (3), let us first have a look at the stationary solutions N for the density matrix. They may occur for the constant pump field amplitude E_p and are characterized by

$$[H(\mathbf{k}), N(\mathbf{k})] = 0. \quad (6)$$

The above equations are usually solved by the method of the Bogoliubov transformation: since H and N are commuting Hermitian matrices, there has to be a unitary transformation which simultaneously diagonalizes H and N . The eigenvalues of H are interpreted as quasiparticle dispersions, in analogy to the Cooper pairs in the BCS theory. Alternatively, the stationary states can be found by the pseudo-spin representation [13, 15, 20], which has the advantage of providing some graphic illustration. As result, the elements of H are given by the coupled equations

$$H_{cv}(\mathbf{k}) = -\mu E_p + \int \frac{d^3\mathbf{k}'}{(2\pi)^3} V(\mathbf{k}-\mathbf{k}') \frac{H_{cv}(\mathbf{k}')}{\sqrt{[H_{cc}(\mathbf{k}') - H_{vv}(\mathbf{k}')]^2 + 4|H_{cv}(\mathbf{k}')|^2}} \quad (7)$$

$$H_{cc}(\mathbf{k}) - H_{vv}(\mathbf{k}) = E_c(\mathbf{k}) - E_v(\mathbf{k}) - \hbar\omega_p \quad (8)$$

$$- \int \frac{d^3\mathbf{k}'}{(2\pi)^3} V(\mathbf{k}-\mathbf{k}') \left[1 - \frac{H_{cc}(\mathbf{k}') - H_{vv}(\mathbf{k}')}{\sqrt{[H_{cc}(\mathbf{k}') - H_{vv}(\mathbf{k}')]^2 + 4|H_{cv}(\mathbf{k}')|^2}} \right].$$

In the absence of an external electromagnetic field E_p , the first equation has the same structure as the gap equation of the BCS theory. Therefore, this nontrivial solution of the semiconductor Bloch equations (3) is referred to as an excitonic insulator. This state is postulated in many theoretical papers on coherently driven semiconductors [8–16]. The fact that the quantity $H_{cc}(\mathbf{k}) - H_{vv}(\mathbf{k})$ has to be calculated self-consistently does not change the result qualitatively, as can be seen from comparing figure 1 of [12] (no self-consistency) and figure 2 of [13] (self-consistency).

Apparently, the only parameters in equations (3) and (8) are the relative exciton mass $m = m_e m_h / (m_e + m_h)$, the pump detuning $E_c(0) - E_v(0) - \hbar\omega_p$, and the coupling to the electromagnetic field μE_p . In order to eliminate unnecessary degrees of freedom, we set the forbidden gap $E_c(0) - E_v(0)$ equal to zero, thus defining $\hbar\omega_p$ relative to the gap. Furthermore, we go over to natural units, $\hbar = m = e^2 / 4\pi\epsilon_0\epsilon = 1$. Then the units of energy, length, and time are $me^4 / (4\pi\epsilon_0\epsilon)^2 \hbar^2$ (twice the exciton binding energy), $4\pi\epsilon_0\epsilon \hbar^2 / me^2$ (the Bohr radius of the exciton), and $\hbar(4\pi\epsilon_0\epsilon)^2 / me^4$. For further simplification, we set $\mu = 1$, which means that E_p has the dimension of an energy and is measured in units of two exciton Rydbergs. For GaAs, the above natural units are 9.34 meV, 11.6 nm, and 70.5 fs; and $\mu E_p = 0.5$ corresponds to a pump intensity of roughly 55 MW cm⁻².

The excitonic insulator is predicted on the basis of equations (7)–(8), which are derived under the assumption that correlation effects can be totally neglected. This is obviously unrealistic and it is worthwhile to study the influence of incoherent processes on the stability of the excitonic insulator. The simplest extension of the model, which is still consistent with the Hartree–Fock approximation is the assumption of exponential dephasing, namely

$$\left. \frac{\partial}{\partial t} N(\mathbf{k}, t) \right|_{\text{corr}} = \begin{pmatrix} 0 & -\gamma \psi(\mathbf{k}, t) \\ -\gamma \psi^*(\mathbf{k}, t) & 0 \end{pmatrix} \quad (9)$$

with a dephasing constant $\gamma > 0$. The quantity $T_2 = 1/\gamma$ is known as the dephasing time. Microscopically, the above ansatz is equivalent to stochastic fluctuations of the band edges in the fast-modulation limit [21], which are attributed to the interaction of the electrons with a phonon bath. This is the reason why the simple T_2 ansatz works very well in practice. Four-wave-mixing experiments have revealed an exponential decay of the polarization over a range of three decades [22].

The diagonal elements of the density matrix are also subjected to incoherent processes, which leads to the relaxation of carriers. These processes are rather inefficient near the band edge. The density of states approaches zero at the band edge and the carrier distributions are nearly thermal, which is unfortunate both for electron–electron and electron–phonon scattering. Furthermore, LO-phonon scattering, the most important scattering mechanism, is forbidden by energy conservation (GaAs: $\hbar\omega_{\text{LO}} = 36$ meV). This is also well established by experiments and quantum-kinetic calculations [23]. Recombination, that is the annihilation of electron-hole pairs, is by order of magnitude slower than dephasing and relaxation.

In order to numerically solve the equation of motion (3), we exploit the spherical symmetry of $H(\mathbf{k})$ and $N(\mathbf{k})$, and the wave number $k = |\mathbf{k}|$ is discretized on a finite interval $k \in [0, k_{\text{max}}]$; $k_{\text{max}} = 65$ with a mesh size $\Delta k = 0.03$. By the sampling theorem, this is equivalent to a discretization in real space with $r_{\text{max}} = 100$ and $\Delta r = 0.05$. The convolution with the Coulomb potential was performed by Fourier transform. Stability is a major issue for the discretization in time. We used the central-difference or leapfrog method,

$$\frac{N(\mathbf{k}, t + \Delta t) - N(\mathbf{k}, t - \Delta t)}{2 \Delta t} = -i [H(\mathbf{k}, t), N(\mathbf{k}, t)] \quad (10)$$

which has previously been successfully tested on nonlinear Schrödinger equations [24].

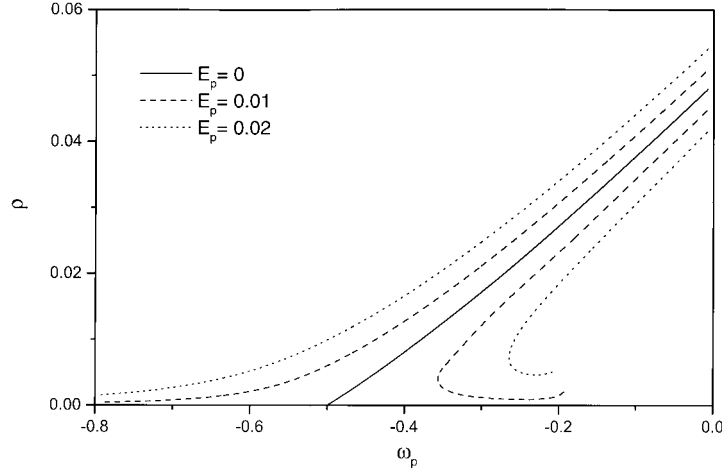


Figure 1. Pair density ρ of the excitonic insulator versus pump frequency ω_p for different pump amplitudes $E_p = 0, 0.01, \text{ and } 0.02$.

The maximum step size, for which the above scheme is stable, is equal the reciprocal norm of the superoperator $[H(k_{\max}), \cdot]$, and can be estimated as $\Delta t_{\max} = 2/k_{\max}^2$.

Accuracy and stability of the numerical algorithm have been verified by (i) extracting the linear absorption from the low-field limit, (ii) reproducing the adiabatic following in the regions below the 1s exciton resonance and between the 1s and 2s exciton resonance, using the excitonic insulator (upper branch in figure 1) as initial condition, and (iii) testing the convergence with respect to the parameters k_{\max} , Δk , and Δt . The numerical result for the linear spectrum was found to be accurate up to $\omega = 10$, which is far above the upper frequency limits of the pulses to be used.

3. Results

To determine the stationary solutions of the semiconductor Bloch equations (3) in the absence of dephasing, we numerically solve the equations (7)–(8). In figure 1, the density ρ is plotted versus the pump frequency ω_p , relatively to the fundamental gap of the semiconductor. Solutions are shown for $E_p = 0$ (solid line), 0.01 (dashed line), and 0.02 (dotted line). Non-trivial solutions (solid line) always start at the exciton resonances $\mathcal{E}_v = -1/2v^2$. For $\omega_p < \mathcal{E}_1 = -1/2$, there is only one solution ρ for each E_p and it holds that $\rho \rightarrow 0$ as $E_p \rightarrow 0$. Thus, no excitonic insulator can be optically generated in this region. For excitation above the first exciton resonance, there are three solutions for a fixed value of E_p . As E_p goes to zero, the upper two branches go over into the non-trivial solution and the lower branch approaches the trivial solution with $\rho = 0$. Although, in principle, an excitonic insulator is possible in the region $\mathcal{E}_1 = -1/2 < \omega_p < \mathcal{E}_2 = -1/8$, this state is unlikely to be realized through adiabatic pulse excitation, because there is no continuous transition from the lower branch to the upper branches of the solution. If the pulse is made intentionally short in order to flip the system over to the upper branch, or the field strength is increased to values where there is only one solution in this region, the adiabaticity is destroyed and the system may not become stationary again. Beyond the second exciton resonance the structure of the solution is going to be even more complicated, because the line of demarcation given by the non-trivial

solution is falling backwards (not shown). Anyway, in this case one cannot speak about the pump pulse being situated between two exciton resonances and only the uppermost branch of the solution is of practical interest. In the end, only a numerical calculation can clarify the relationship between the stationary solutions of equations (7)–(8) and the time-dependent solutions of equation (3).

For the time-dependent solution of equation (3) we use a Gaussian pulse envelope of the form

$$E_p(t) = E_{\max} \exp(-t^2/2\sigma^2) \quad (11)$$

which should be close to ideal, because the product of frequency bandwidth and time duration is at minimum. We believe that this assumption does not limit the generality of the results. Anyway, the experimental setup does not allow the design of arbitrary laser pulses.

The signatures of an excitonic insulator would be (i) a stationary density and (ii) a constant polarization since, by means of the rotating-wave approximation, the pump frequency is incorporated in the band dispersions (3).

We focus on resonant excitation, $\omega_p > \mathcal{E}_1$, where figure 1 proposes non-trivial solutions. The semiconductor Bloch equations (3) were solved for various parameters $-0.3 \leq \omega_p \leq +0.3$, $0.005 \leq E_p \leq 0.4$, $1.0 \leq \sigma \leq 16$, without dephasing ($\gamma = 0$) and with dephasing ($\gamma = 0.1$). Out of the multitude of data, only a few curves can be shown.

First, we study the collision-free case with $\gamma = 0$. In figure 2(a), the time evolution of the pair density ρ is shown for fixed pump frequency $\omega_p = -0.3$, fixed pulse length $\sigma = 4$, and different pump amplitudes $E_{\max} = 0.005, 0.01, 0.02$, and 0.04 . In each case, a finite density is created and the density always remains constant for times much larger than the pulse length. However, a look at the polarization reveals that the solutions are not at all stationary. In figure 2(b), the functions $\text{Re } \psi(k=0, t)$ (solid line) and $\text{Re } \psi(k=1, t)$ (dashed line) are plotted versus t for the same parameters. The oscillations are completely irregular and strongly dependent on the pump amplitude. There is hardly any correlation between the oscillations at different wave numbers. The density, which would correspond to an excitonic insulator, $\rho = 0.0172$ (figure 1), is obtained for $E_{\max} = 0.0262$. Even if E_{\max} is tuned such that the stationary density equals the density of the excitonic insulator, the polarization shows strong beats at any value of k (not shown).

Figure 3(a) shows the density versus time for a much longer pulse $\sigma = 16$. The other parameters are the same as in figure 2. The pump intensity $I(t) \propto |E(t)|^2$ is shown as a dotted curve. Due to the large pulse length and the neglect of dephasing, the system may follow the pump pulse adiabatically. This is possible only as long as a lowest branch of the stationary solution at the given pump frequency exists (figure 1). Indeed, in those cases ($E_{\max} \leq 0.01$) we observe adiabatic following of the density. For larger pulse amplitudes, the density rapidly increases and exceeds the density of the excitonic insulator. The time-dependent polarization is shown in figure 3(b). The polarization $\psi(k, t)$ is completely irregular for each k . An exception is a pump amplitude $E_{\max} = 0.02$, which will be discussed later in some more detail. Even for those pulses where we observed adiabatic following of the density, there is still some polarization left after the end of the pulse. Again, we find that, when the field is chosen such that the stationary density equals the density of the excitonic insulator ($E_{\max} = 0.0167$, not shown), the polarization is not stationary. This is consistent with figure 1, where a quasi-static transition into an excitonic insulator is forbidden by the topology of the solution.

We have also studied the time evolution of density and polarization for different pump frequencies and pulse lengths. The result is essentially the same: while the density always approaches a stationary value, the components of the polarization show irregular oscillations, which are sensitive to the parameters of the pump pulse. Figure 4 shows the time dependence

of density (a) and polarization (b) for various pump frequencies $\omega_p = -0.3, -0.1, +0.1$ and $+0.3$. The other pulse parameters are fixed at $E_{\max} = 0.02$ and $\sigma = 8$. For $\omega_p = -0.3$, the stationary density is above the density of the excitonic insulator at $\omega_p = -0.3$. For the higher pump frequencies, the stationary density remains below the excitonic-insulator density at those pump frequencies. Moreover, the function $\rho(t)$ is virtually the same for all pump frequencies $\omega_p \geq -0.1$ and there is no difference between pump frequencies below and above the continuum edge. This weak dependence on the pump frequency is also observed for other values of the amplitude and pulse length, independent of whether the stationary density lies below or above the excitonic-insulator density (not shown), and can be explained as follows: the 1s exciton, by virtue of its large oscillator strength, most strongly contributes to the density of the excitonic insulator. If the spectral overlap of the pump pulse with the 1s exciton is negligible, the response of the system is essentially non-resonant. This is also seen in the polarization (figure 4 (b)): for excitation which is non-resonant to the 1s exciton, different components of the polarization oscillate with completely different frequencies, as one would expect from interaction-free particles.

It is worthwhile to revisit the case where different k -components of the polarization oscillate with about the same frequency ($\omega_p = -0.3$; $\sigma = 16$; $E_{\max} = 0.02$, second from the top in figure 3 (b)). We studied the polarization for pump amplitudes in the neighborhood of $E_{\max} = 0.02$ in some more detail and found that for each wave number the polarization shows oscillations proportional to $\exp(-2\pi it/T)$ with a period $T = 30$. Since we operate in the rotating frame, the actual frequency, relative to the band edge is $\omega = \omega_p + 2\pi/T = -0.09$. For a slightly smaller amplitude, $E_{\max} = 0.0195$, the stationary density $\rho = 0.0389$ is equal to the density of the excitonic insulator at $\omega = -0.09$. This is the non-stationary excitonic insulator, as previously described by Östreich and Schönhammer [15]: depending on the energy transmitted by the pump pulse, the system may exhibit collective oscillations with a frequency that differs from the pump frequency. In figure 5, real part (solid line) and imaginary part (dashed line) of the polarization are shown for different wave numbers $k = 0, 1$, and 3.

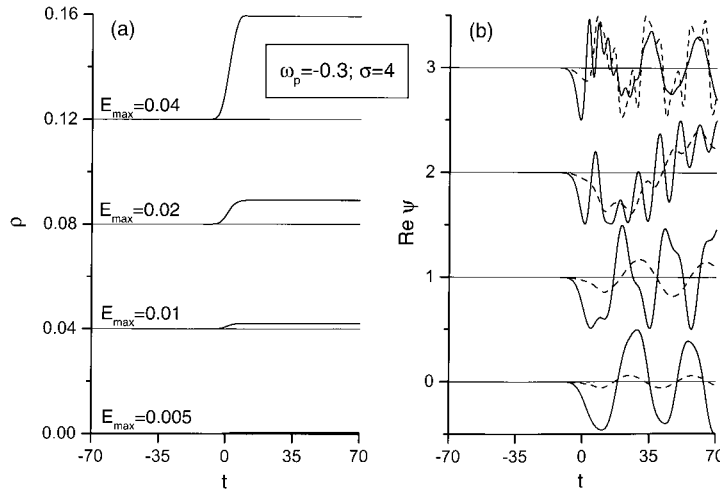


Figure 2. Time evolution of the pair density and polarization for $\omega_p = -0.3$, $\sigma = 4$, $\gamma = 0$, and different pulse amplitudes $E_{\max} = 0.005, 0.01, 0.02$, and 0.04 . (a) ρ versus t . (b) $\text{Re } \psi(k, t)$ versus t for $k = 0$ (solid line) and $k = 1$ (dashed line).

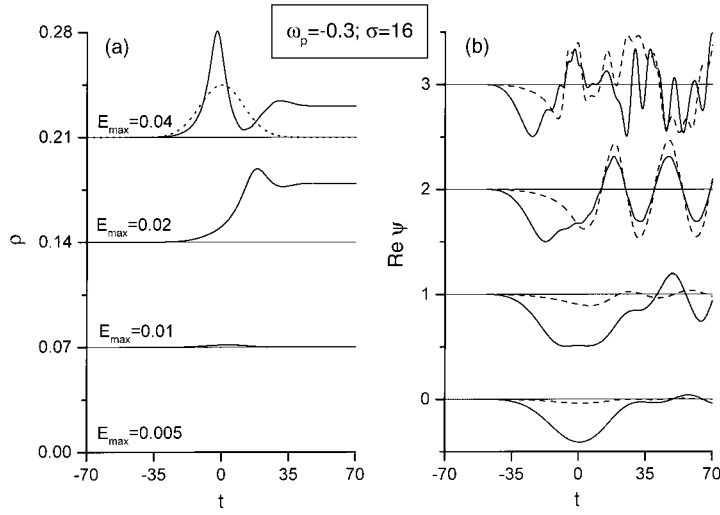


Figure 3. Time evolution of the pair density and polarization for $\omega_p = -0.3$, $\sigma = 16$, $\gamma = 0$, and different pulse amplitudes $E_{\max} = 0.005, 0.01, 0.02$, and 0.04 . (a) ρ versus t (solid line) and pump intensity $I(t) \propto |E(t)|^2$. (b) $\text{Re } \psi(k, t)$ vs. t for $k = 0$ (solid line) and $k = 1$ (dashed line).

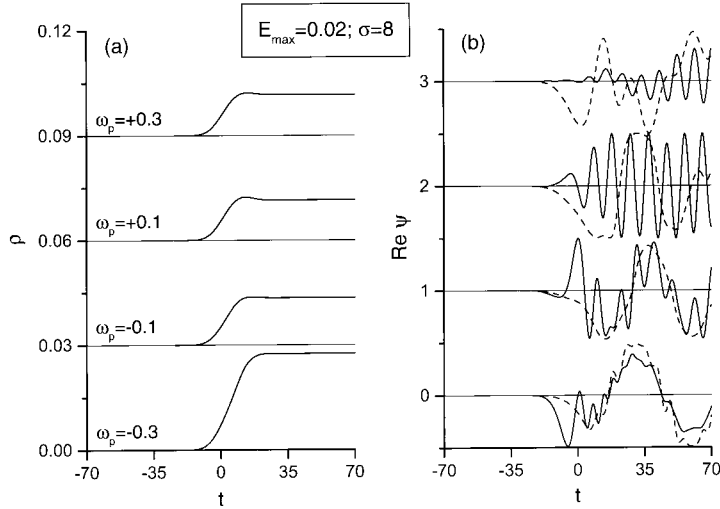


Figure 4. Time evolution of the pair density and polarization for $E_{\max} = 0.02$, $\sigma = 8$, $\gamma = 0$, and different pump frequencies $\omega_p = -0.3, -0.1, +0.1$, and $+0.3$. (a) ρ vs. t . (b) $\text{Re } \psi(k, t)$ vs. t for $k = 0$ (solid line) and $k = 1$ (dashed line).

The pulse amplitude is $E_{\max} = 0.0195$ and the other parameters are the same as in figure 3. In the long-time limit, the components of the polarization show oscillations that are almost sinusoidal, and have the same period $T = 30$ for each k . Moreover, like the excitonic insulator, the complex phase is the same for each $\psi(k, t)$.

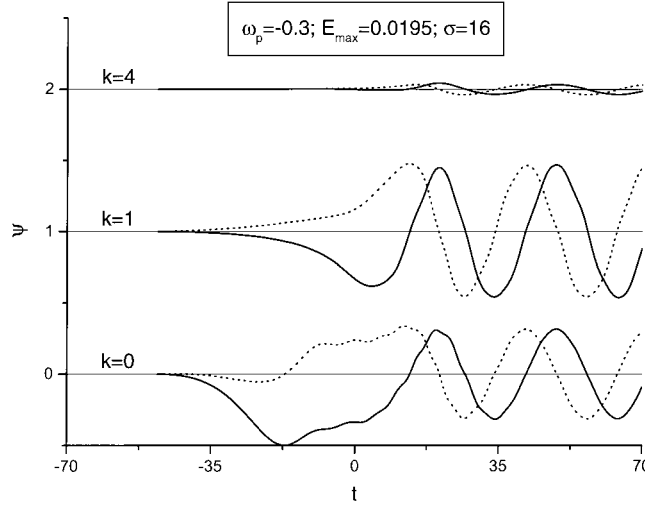


Figure 5. Polarization $\text{Re } \psi(k, t)$ (solid line) and $\text{Im } \psi(k, t)$ (dotted line) versus time t at different wave numbers $k = 0, 1, 3$ for a non-stationary excitonic insulator. The parameters of the pulse are $\omega_p = -0.3$, $\sigma = 4$, and $E_{\text{max}} = 0.0195$.

At this point we conclude that, if dephasing is neglected, the creation of an excitonic insulator in the strict sense is not possible by pulse excitation. On the other hand, the transition into a non-stationary excitonic insulator may occur for certain parameters.

Now we turn to the situation, when the polarization is subjected to dephasing. We will specifically address two questions: (i) provided the system is prepared in the state of an excitonic insulator, what is the time scale for the relaxation into the normal state? and (ii) how is the creation of the excitonic insulator itself affected by the dephasing?

The answer to question (i) is that the lifetime of an excitonic insulator equals T_2 . For a proof, we assume that the semiconductor is in the state of an excitonic insulator at $t = 0$ so that $[H(\mathbf{k}, 0), N(\mathbf{k}, 0)] = 0$. From equations (3) and (9) it follows immediately that

$$N(\mathbf{k}, t) = \begin{pmatrix} N_{cc}(\mathbf{k}, 0) & N_{cv}(\mathbf{k}, 0) e^{-\gamma t} \\ N_{vc}(\mathbf{k}, 0) e^{-\gamma t} & N_{vv}(\mathbf{k}, 0) \end{pmatrix} \text{ for } t \geq 0. \quad (12)$$

Thus the components of the polarization decay exponentially, whereas—in this approximation—the diagonal elements of the density matrix remain unchanged. This result also applies to the non-stationary excitonic insulator, which is equivalent to an excitonic insulator at a frequency that differs from the pulse frequency. While this result is not encouraging, at least it leaves us with the hope of observing an excitonic insulator within a time span of T_2 (e.g., when the delay between pump and probe pulse is smaller than T_2).

To answer question (ii), we numerically solve the semiconductor Bloch equations (3), with a dephasing term according to equation (9) and a dephasing constant $\gamma = 0.1$ (GaAs: $T_2 = 705$ fs). For comparison with the collision-free case, we use the same parameters as in figure 3: $\omega_p = -0.3$, $\sigma = 16$, and $E_{\text{max}} = 0.005, 0.01, 0.02, 0.04$. The results for the density and polarization are shown in figure 6(a) and (b), respectively. For low pump intensities, no adiabatic following of the density is observed. Instead, the density increases monotonically to a finite value. In contrast to the collision-free case, the polarization always drops to zero. Yet the components of the polarization are very different from the collision-free result, multiplied with

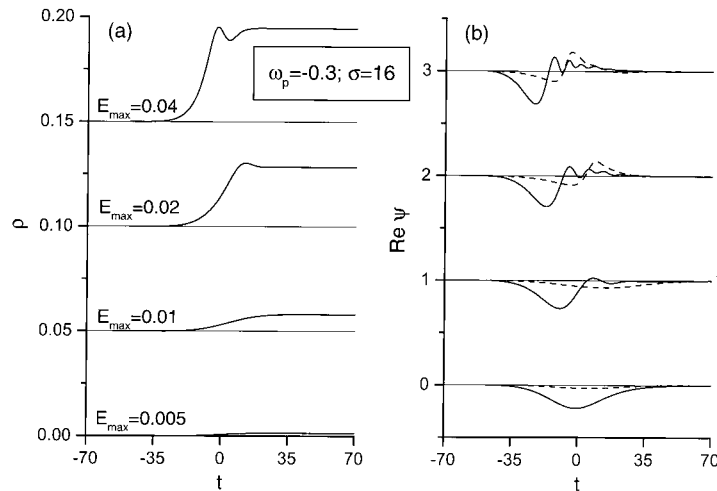


Figure 6. Time evolution of the pair density and polarization in the presence of dephasing with $\gamma = 0.1$. The other parameters are the same as in figure 3.

$\exp(-\gamma t)$, as suggested by equation (12). This is prevalent for $k = 1$, where the polarization shows much less oscillations than in the collision-free case. Also, for $E_{\max} = 0.02$, which is close to the non-stationary excitonic insulator in the collision-free case, there is no correlation whatsoever between $\psi(k = 0, t)$ and $\psi(k = 1, t)$. We also checked pulse amplitudes in the vicinity of $E_{\max} = 0.02$ and came to the same result. In conclusion, a finite dephasing time does not only destroy an excitonic insulator, but also prevents the buildup of the excitonic insulator by pulse excitation.

4. Summary

In summary, we have provided an accurate solution of the semiconductor Bloch equations, including continuum states, without dephasing and in the framework of linear exponential dephasing. The time-dependent pair density has been compared with the stationary solution (gap equation) for a large variety of parameters.

For no set of parameters was an excitonic insulator directly obtained by pulse excitation. In the collision-free approximation, for certain parameters, a non-stationary excitonic insulator is obtained.

The effect of dephasing is devastating. If a hypothetical excitonic insulator is exposed to dephasing, the non-diagonal elements of the density matrix decay exponentially with characteristic time T_2 . If the dephasing is taken into account from the very beginning, no excitonic insulator is created. Thus we conclude that the excitonic insulator is unlikely to be realized by means of coherent pulse excitation.

Acknowledgment

This work was supported by the Thüringer Ministerium für Wissenschaft, Forschung und Kultur.

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