

Coherent optics for pumping near the absorption edge

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The optical properties of a semiconductor pumped near the 1s exciton resonance are studied. The equation of motion for the density matrix in the Hartree-Fock approximation is traced back to the Bloch equation, well known from two-level systems. The essential difference, however, is the coupling of different wave vectors via the Coulomb potential. Principal features of the different stationary solutions are discussed. For a closer inspection of stability, we calculate the response to a smooth switchon of the pump field. For pump frequencies above the 1s exciton, the system follows first adiabatically the pulse shape, showing a blueshift and a slight increase of the oscillator strength as in the off-resonant case. The system jumps later into a second solution characterized by optical gain below the pump frequency, related to complete saturation (excitonic insulator). The major objective of our paper is to provide a numerical solution to the equation of motion in the Hartree-Fock approximation and to compare the results to those obtained by stationary considerations.

I. INTRODUCTION

The recent developments of subpicosecond spectroscopy have allowed the observation of new coherent phenomena in semiconductors. Experiments under non-resonant excitation have revealed light-induced changes of excitonic energies and corresponding oscillator strengths.¹⁻⁸ A microscopic description of this phenomenon, known as the optical Stark effect, has been performed by Schmitt-Rink and co-workers.^{9,10} In this context the virtually excited carriers, i.e., coherently driven electrons and holes which experience no real collisions, play an important role; they are also responsible for changes of the screening properties.^{11,12} Two-particle states have to be considered if changes in the excitonic absorption are of interest. Numerical results of the corresponding theory^{9,10} have been obtained by Ell *et al.*^{13,14} and Schäfer, Schuldt, and Binder.¹⁵ Analytically the optical absorption of excitons in the coherent regime can be traced back to a generalized Elliot formula.¹⁶ A possible influence of biexcitons is considered in a series of papers.¹⁷⁻²⁰

An adequate description of these nonequilibrium phe-

nomena is possible using the Keldysh technique.^{21,22} Most of the concepts developed in the context of the optical Stark effect have their origin in the theory of two-level atoms.²³⁻²⁵ However, the inclusion of Coulomb effects leads to an inhomogeneous gap equation. Its homogeneous version is well known from the theory of superconductivity. An interesting question in this context is the existence and observability of an excitonic insulator.²⁶⁻³⁰

A stationary consideration of the equation of motion for the density matrix in the pump-frequency region above the 1s exciton position leads to different solutions;^{16,29} the physical relevance in each case, however, has to be examined by solving the time-dependent equation.

II. INHOMOGENEOUS GAP EQUATION

We consider a two-band model semiconductor interacting with a classical quasimonochromatic pump field. The interaction between light and matter is treated within the dipole approximation. The dynamics of the system is thus determined by a Hamiltonian of the form³¹⁻³³

$$H = \sum_{\mathbf{k}} \sum_{i=1}^2 E_i(\mathbf{k}) c_{i\mathbf{k}}^\dagger(t) c_{i\mathbf{k}}(t) - \sum_{\mathbf{k}} \sum_{\substack{i,j=1 \\ i \neq j}}^2 \mu_{ij}(\mathbf{k}) \cdot \mathbf{E}(t) c_{i\mathbf{k}}^\dagger(t) c_{j\mathbf{k}}(t) + \frac{1}{2} \frac{1}{\Omega} \sum_{\mathbf{k}, \mathbf{k}' \mathbf{q}} \sum_{i,j=1}^2 V(\mathbf{q}) c_{i\mathbf{k}-\mathbf{q}}^\dagger(t) c_{j\mathbf{k}'+\mathbf{q}}^\dagger(t) c_{j\mathbf{k}}(t) c_{i\mathbf{k}}(t). \quad (1)$$

Here $c_{i\mathbf{k}}^\dagger$ ($c_{i\mathbf{k}}$) represents the creation (annihilation) operator of an electron in the Bloch state $|i, \mathbf{k}\rangle$ ($i=1,2$) with energy $E_i(\mathbf{k})$. The pump field $\mathbf{E}(t) = \mathbf{E}_p(t) \exp(-i\omega_p t) + \text{c.c.}$ is considered to be nearly monochromatic, i.e., the

pump-field amplitude $\mathbf{E}_p(t)$ is assumed to be a slowly varying function of time t . The Coulomb potential $V(\mathbf{q}) = e^2 / \epsilon_0 \epsilon_b q^2$ is responsible for the electron-electron interaction, ϵ_b being the dielectric constant of the semi-

conductor. Ω denotes the crystal volume. The spin of the electrons is not explicitly taken into account and appears occasionally as a factor of 2.

The one-particle properties are governed by a set of Green's functions defined by the expectation value

$$G_{ijAB}(\mathbf{k}, t_1, t_2) = \frac{1}{i\hbar} \langle T_{AB} c_i(\mathbf{k}, t_1) c_j^\dagger(\mathbf{k}, t_2) \rangle. \quad (2)$$

The indices A and B at the time-ordering operator T refer to different branches of the Keldysh contour,^{21,34,35} taking into account that spectral and kinetic properties cannot be traced back to one type of Green's functions as in the equilibrium case.^{33,36,37}

For the density matrix

$$n_{ij}(\mathbf{k}, t) = -i\hbar G_{-+ij}(\mathbf{k}, t, t) \quad (3)$$

we obtain the equation of motion in Hartree-Fock approximation:^{33,36,38-40}

$$i\hbar \frac{\partial}{\partial t} n(\mathbf{k}, t) = [E(\mathbf{k}, t), n(\mathbf{k}, t)],$$

$$E(\mathbf{k}, t) = \begin{pmatrix} E_c(\mathbf{k}) & F(t) \\ F^*(t) & E_v(\mathbf{k}) \end{pmatrix} - \frac{1}{\Omega} \sum_{\mathbf{q}} V(\mathbf{k}-\mathbf{q}) [n(\mathbf{q}, t) - n^{(0)}(\mathbf{q})], \quad (4)$$

$$F(t) = -\boldsymbol{\mu} \cdot \mathbf{E}(t),$$

which is identical to that obtained by Schmitt-Rink and co-workers.^{9,10} $n^{(0)}$ represents the density matrix before excitation.

The appearance of single-particle energies and Hartree-Fock self-energy as a unique energy matrix E in the equation of motion (4) gives rise to an interpretation of the semiconductor as a system of noninteracting particles with renormalized energies.²⁹ However, the presence of nondiagonal elements and its time dependence via the pump-field amplitude produces some complications. (i) In the stationary case, the Hamiltonian and the Green's function can immediately be diagonalized by a Bogoliubov transformation, but the resulting coherent ground state cannot trivially be determined by a stationary consideration since it still contains degrees of freedom due to the Coulomb interaction. (ii) Apart from this fact the validity of the collision-free limit is by no means fully clarified; of special interest in this context is the relation between the coherent ground state postulated and the completely relaxed state.^{10,26,29} (iii) The coupling of states with different wave vectors leads to a complicated dynamics^{41,42} even if correlations outside the Hartree-Fock approximation are completely neglected. The be-

$$E_{12}(\mathbf{k}) = F + \frac{1}{\Omega} \sum_{\mathbf{q}} V(\mathbf{k}-\mathbf{q}) s(\mathbf{q}) \frac{E_{12}(\mathbf{q})}{\sqrt{[E_{11}(\mathbf{q}) - E_{22}(\mathbf{q})]^2 + 4|E_{12}(\mathbf{q})|^2}}, \quad (9)$$

which, however, cannot be solved separately since the quantity $E_{11}(\mathbf{q}) - E_{22}(\mathbf{q})$ in the denominator of the right-hand side has to be determined self-consistently leading to

$$E_{11}(\mathbf{k}) - E_{22}(\mathbf{k}) = E_c(\mathbf{k}) - E_v(\mathbf{k}) - \hbar\omega_p - \frac{1}{\Omega} \sum_{\mathbf{q}} V(\mathbf{k}-\mathbf{q}) \left[\frac{1}{2} - \frac{1}{2} s(\mathbf{q}) \frac{E_{11}(\mathbf{q}) - E_{22}(\mathbf{q})}{\sqrt{[E_{11}(\mathbf{q}) - E_{22}(\mathbf{q})]^2 + 4|E_{12}(\mathbf{q})|^2}} \right]. \quad (10)$$

havior depends strongly on the pulse shape of the electromagnetic field. Therefore, careful numerical analyses are necessary.

To cast Eq. (4) into a more intuitive form we regard the Hermitian matrices E , n and (4) as spinor representations of four vectors \underline{E} and \underline{n} given by

$$E = \underline{E}^i \sigma_i, \quad n = \underline{n}^i \sigma_i, \quad (5)$$

$$\underline{E} = \begin{pmatrix} \frac{1}{2}(E_{11} + E_{22}) \\ \frac{1}{2}(E_{11} - E_{22}) \\ \text{Re}E_{12} \\ \text{Im}E_{12} \end{pmatrix}, \quad \underline{n} = \begin{pmatrix} \frac{1}{2}(n_{11} + n_{22}) \\ \frac{1}{2}(n_{11} - n_{22}) \\ \text{Re}n_{12} \\ \text{Im}n_{12} \end{pmatrix},$$

where the σ_i are Pauli matrices. The conservation of the trace in (4) corresponds to the conservation of the "temporal" components \underline{n}^0 in (5), the resulting equation for the "spatial" components being

$$i\hbar \frac{\partial}{\partial t} \underline{n} = -2\underline{E} \times \underline{n}, \quad (6)$$

which represents the Bloch equation, well known from theory of two-level systems. The essential difference, however, is that \underline{E} itself depends on \underline{n} via the Coulomb coupling (4). In this context the terms $n_{11} - n_{22}$ and n_{12} are usually referred to as inversion and polarization, respectively. The gap function $E_{12}(\mathbf{k})$ as an effective field plays an important role in the theory of superconductivity^{43,44} and is of special interest, too, for the description of the optical Stark effect.^{9,10,26,29,32}

To construct stationary solutions of Eq. (6) we have to claim that the vectors \underline{E} and \underline{n} are parallel. Without loss of generality we suppose that the state before excitation is characterized by an empty conduction and a filled valence band. As a consequence of conservation of the determinant in Eq. (4) the length of \underline{n} is conserved for each \mathbf{k} leading to

$$\underline{n}(\mathbf{k}) = -\frac{1}{2} s(\mathbf{k}) \frac{\underline{E}(\mathbf{k})}{|\underline{E}(\mathbf{k})|}, \quad s \in \{-1, 1\}. \quad (7)$$

The explicit expressions for the elements of the density matrix are

$$n_{11}(\mathbf{k}) = \frac{1}{2} - \frac{1}{2} s(\mathbf{k}) \frac{E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})}{\sqrt{[E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})]^2 + 4|E_{12}(\mathbf{k})|^2}},$$

$$n_{22}(\mathbf{k}) = 1 - n_{11}(\mathbf{k}), \quad (8)$$

$$n_{12}(\mathbf{k}) = -s(\mathbf{k}) \frac{E_{12}(\mathbf{k})}{\sqrt{[E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})]^2 + 4|E_{12}(\mathbf{k})|^2}},$$

Introducing Eq. (8) into the definition of the energy matrix (4) we obtain the inhomogeneous gap equation

Considering the structure of Eq. (7) one has to expect that the physically relevant solutions are characterized by a continuous function $s(\mathbf{k})$ which leads to $s(\mathbf{k}) \equiv 1$. The choice $s(\mathbf{k}) \equiv -1$ would correspond to a filled conduction and an empty valence band. On the other hand, having in mind the case of resonant excitation but zero pump intensity, one could suppose that $s(\mathbf{k}) = \text{sgn}[E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})]$ leading to $n_{11}(\mathbf{k}) \leq \frac{1}{2}$ and a kink at the resonance.

Assuming stationary from the outset, the sgn function $s(\mathbf{k})$ remains undetermined, as already discussed in Ref. 10. By solving the differential equation (6) numerically we will clarify this point in the present paper. As shown below we find that $s(\mathbf{k}) \equiv 1$ holds already in the Hartree-Fock approximation without taking into account scattering or relaxation.

In the further treatment all quantities are expressed in excitonic units Rydberg energy \mathcal{R} and Bohr radius a_B , respectively. Solutions for different detunings are shown in Fig. 1. The pulse switchon is modeled by using, for the field amplitude $F(t)$, the error function with rise time τ . For slowly increasing amplitudes (large values of τ) the

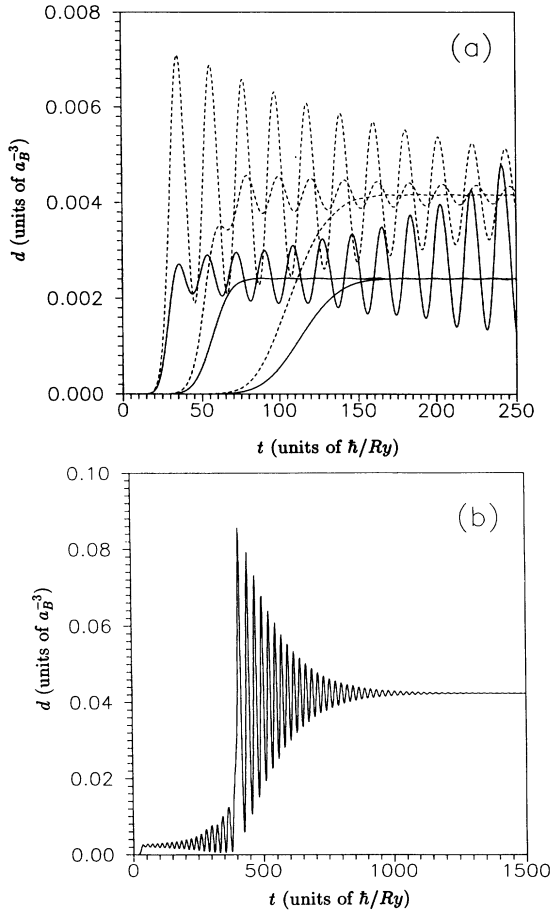


FIG. 1. (a) Density of electron-hole pairs d vs time t for non-resonant ($\hbar\omega_p - E_g = -1.2$, dashed line) and resonant excitation ($\hbar\omega_p - E_g = -0.6$, solid line) and different pulse shapes: $\tau = 16.7, 33.3, 66.7, \lambda = 0.02$; (b) transition from the metastable into a stable regime, $\tau = 16.7, \lambda = 0.02$.

density follows adiabatically the pump intensity whereas for small values of τ Rabi oscillations appear which vanish with time for $\hbar\omega_p - E_g < -1$ as a result of interference of states with different wave vectors. For pump frequencies above the exciton resonance ($-1 < \hbar\omega_p - E_g < 0$) the density follows at first the pulse shape, but occurring Rabi oscillations will be amplified. Therefore, we will apply the term “metastable solution” to this phenomenon. Finally, the system will turn into a stable regime, characterized by a drastic Coulomb-enhanced gap function $E_{12}(\mathbf{k})$. Even if the pump field is switched off again (not shown here) a final density and polarization remain. This phenomenon is closely related to the excitonic insulator²⁸ at $T=0$ with a chemical potential $\mu = \hbar\omega_p$. In each case we have found that $s(\mathbf{k}) \equiv 1$, astonishing in that in the nominal off-resonant situation with $-1 < \hbar\omega_p - E_g < 0$ the bands may already overlap, i.e., $E_{11}(0) - E_{22}(0) < 0$. However, the choice of the function $s(\mathbf{k})$ is of no influence on the principal features of the solution. The same is true for the approximate treatment²⁶ of the gap equation (9) by neglecting the Coulomb contribution in the diagonal elements of the energy matrix (10). The energy of the excitonic insulator can be derived from Eq. (1) using Eqs. (4) and (8). We obtain

$$\Delta E = 2 \left[\hbar\omega_p \sum_{\mathbf{k}} n_{11}(\mathbf{k}) - \sum_{\mathbf{k}} \left\{ \frac{1}{2} \sqrt{[E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})]^2 + 4|E_{12}(\mathbf{k})|^2} - \frac{1}{2}[E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})] \right\} \right] \quad (11)$$

if the total energy before excitation is scaled to zero. This result can be simply understood: The first term represents the energy of the virtually excited electron-hole pairs, the second term results from the renormalization of the (occupied) valence band.

The density of excited electron-hole pairs versus detuning^{26,29} is plotted in Fig. 2. For excitations above the 1s

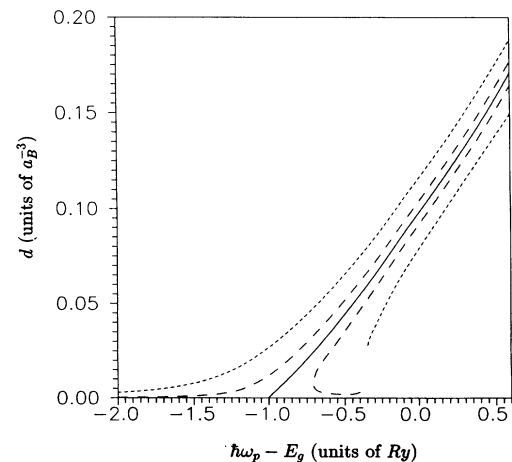


FIG. 2. Density d vs detuning $\hbar\omega_p - E_g$ for different intensities corresponding to $F=0$ (solid line), $F=0.02$ (dashed line), and $F=0.06$ (dotted line).

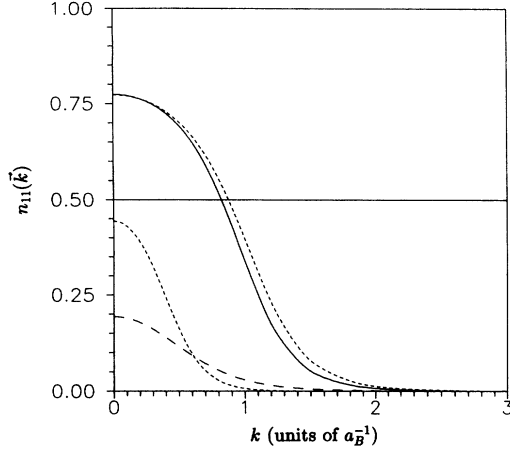


FIG. 3. Distribution function $n_{11}(\mathbf{k})$ for $\hbar\omega_p = -1.2$ (dashed line) and -0.6 (metastable solution, dotted line) for $F=0.02$. The distribution for the excitonic insulator (no excitation, $F=0$) is shown as solid line.

exciton energy we found three solutions. As deduced from the time-dependent calculation, a metastable (lowest density), unstable (medium density), and stable (highest density) solution are observed. The unstable solution is of no physical relevance but could be found by iterating Eq. (7). The stable solution depends weakly on the exter-

nal field and retains a residual density and polarization if the external field is switched off again. It corresponds thus to a saturated state. It is worth mentioning that for both the metastable and the unstable solution the external and internal fields partially compensate each other, i.e., $\arg(F) = \arg[n_{12}(\mathbf{k})]$ for each \mathbf{k} , whereas the stable solution is characterized by $\arg(F) = -\arg[n_{12}(\mathbf{k})]$. A possible additional pair of solutions above the $2s$ exciton resonance could not clearly be stated here because of numerical difficulties in this region.

Figure 3 shows the distribution function $n_{11}(\mathbf{k})$ for zero and nonzero pump field amplitude and pump frequencies below and above the $1s$ exciton energy. The stable solution is characterized by a \mathbf{k} region where $n_{11}(\mathbf{k}) \geq \frac{1}{2}$, which is equivalent to $E_{22}(\mathbf{k}) > E_{11}(\mathbf{k})$.

III. OPTICAL ABSORPTION

The dielectric function describes the linear response of a system to a weak test beam. In the Hartree-Fock approximation the test-field-induced polarization $\delta n_{cv}(\mathbf{k})$ can be immediately derived from the equation of motion (4) imposing a weak perturbation $\delta\mathbf{E}$ oscillating with frequency ω on the pump field \mathbf{E}_p .^{10,15} In the stationary case an alternate way is to solve the Bethe-Salpeter equation for the polarization function.^{36,37} Application of a Bogoliubov transformation leads to

$$\chi(\omega) = -2|\mu|^2 \frac{1}{\Omega} \sum_{\mathbf{k}_1, \mathbf{k}_2} \Phi(\mathbf{k}_1) \cdot \underline{H}^{-1}(\mathbf{k}_1, \mathbf{k}_2, \omega - \omega_p + i\varepsilon) \cdot \Phi(\mathbf{k}_2),$$

$$\underline{H}(\mathbf{k}, \mathbf{k}', z) = \delta_{\mathbf{k}\mathbf{k}'} \hbar \begin{bmatrix} \omega_1 - \omega_2 - z & 0 \\ 0 & \omega_1 - \omega_2 + z \end{bmatrix} - \frac{1}{\Omega} V(\mathbf{k} - \mathbf{k}') \begin{bmatrix} \alpha\alpha' + \beta\beta' + 2\gamma\gamma'^* & 2\gamma\gamma'^* - \alpha\beta' - \beta\alpha' \\ 2\gamma\gamma'^* - \alpha\beta' - \beta\alpha' & \alpha\alpha' + \beta\beta' + 2\gamma\gamma'^* \end{bmatrix},$$

$$\Phi(\mathbf{k}) = \begin{bmatrix} \alpha(\mathbf{k}) \\ -\beta(\mathbf{k}) \end{bmatrix},$$

$$\alpha(\mathbf{k}) = \frac{1}{2} + \frac{1}{2} \frac{E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})}{\sqrt{[E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})]^2 + 4|E_{12}(\mathbf{k})|^2}}, \quad (12)$$

$$\beta(\mathbf{k}) = 1 - \alpha(\mathbf{k}), \quad \gamma(\mathbf{k}) = \frac{E_{12}(\mathbf{k})}{\sqrt{[E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})]^2 + 4|E_{12}(\mathbf{k})|^2}},$$

$$\hbar\omega_{1/2} = \frac{1}{2}[E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})] \pm \frac{1}{2}\sqrt{[E_{11}(\mathbf{k}) - E_{22}(\mathbf{k})]^2 + 4|E_{12}(\mathbf{k})|^2},$$

for $s(\mathbf{k}) \equiv 1$. For further details see Ref. 16. Equation (12) can be looked upon as a generalization of the well-known Elliot formula.⁴⁵ The energy difference $\hbar\omega_1 - \hbar\omega_2$ corresponds to the renormalized band structure, and the modifications of the Coulomb potential result from Pauli blocking due to virtually excited carriers.

The imaginary part of the interband dielectric func-

tion, responsible for the optical absorption, is displayed in Fig. 4 for zero and nonzero intensity and different values of detuning. A final line width $\varepsilon=0.1$ is introduced artificially. For $\hbar\omega_p - E_g < -1$ the well-known results^{14,15,46} are reproduced: The coherent excitation produces a blueshift and a slightly increasing oscillator strength. In the case $-1 < \hbar\omega_p - E_g < 0$ we have calcu-

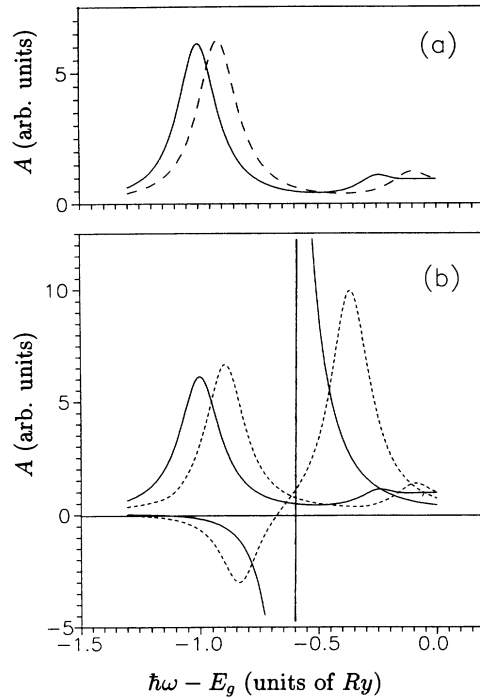


FIG. 4. Optical absorption A vs frequency ω for nonresonant (a) and resonant (b) excitation corresponding to the distribution functions in Fig. 3.

lated curves corresponding to the different stationary states (cf. Figs. 2 and 3). The metastable solution behaves like the off-resonant case (blueshift for increasing pump intensity). The corresponding Stark shift has been calculated already in Ref. 47. The excitonic insulator is characterized by a pole just at the pump frequency ω_p , being equivalent to the Mahan exciton.⁴⁸ As in the off-resonant case the existence of a gain region is a result of

three-photon processes.^{10,46}

We emphasize that the order of magnitude of the linewidth introduced corresponds to experimental data. This seems to be contradictory to the complete neglecting of dephasing and relaxation. This assumption, however, is not crucial. As further investigations have shown,⁴⁹ both metastable and stable solution (cf. Fig. 1) may occur at a realistic time scale (a few picoseconds for GaAs).

IV. SUMMARY

The equation of motion in Hartree-Fock approximation (4) could be rewritten into the optical Bloch equation (5) which can be in a simple way interpreted geometricaly. The structure is just the same as for the two-level system; however the "axis" \underline{E} itself depends on \underline{n} . The numerical solution showed that, in any case, the $s(\mathbf{k})$ introduced in Eq. (7) comes out to be $s(\mathbf{k}) \equiv 1$ even in the case of resonant excitation where the distribution function $n_{11}(\mathbf{k})$ exceeds the value $\frac{1}{2}$. This is in accordance with the argument that physical situations should be characterized by continuous functions (density and polarization over \mathbf{k}). Although the excitonic insulator is stable, i.e., occurring Rabi oscillations disappear, it cannot be reached adiabatically.

The optical spectrum of the excitonic insulator is characterized by a pole at the position of the pump frequency. At finite intensities oscillator strength is shifted away from the pump frequency (opening of the laser-induced gap). The gain region in the optical spectrum has the same origin as the three-photon process at negative detuning.

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