

Rabi oscillations in semiconductor multiwave mixing response

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We studied the semiconductor response with respect to high intensity resonant excitation on short-time scale when the contribution of the Fermi statistics of the electrons and holes prevails. We studied both the single- and double-pulse excitations. For the latter case we considered the time evolution of the multiwave mixing exciton polarization. The main difference between the excitation by a single pulse or by two noncollinear pulses is that the Rabi oscillations of the multiwave mixing response are characterized by two harmonics. Analyzing the operator dynamics governed by the external excitation we found that there are three invariant spin classes, which do not mix with the evolution of the system. Two classes correspond to the bright exciton states and one contains all dark states. We found that the dynamics of the classes are described by six frequencies and the Rabi frequencies are only two of them (one for each bright class). We discuss the effect of the dispersion of the electrons and holes and the Coulomb interaction describing the semiconductor by the semiconductor Bloch equation (SBE). We show that if initially the system is in the ground state then the SBE preserves the invariant spin classes, thus proving the absence of the dark excitons in the framework of this description. We found that due to the mass difference between holes of different kind additional Rabi frequencies, two of those present in the operator dynamics should appear in the evolution of the exciton polarization.

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I. INTRODUCTION

One of the main tools of probing the complex character of the many-body correlations and interactions in semiconductors is the multiwave mixing (MWM) response. The MWM polarization in optically excited semiconductors produces the signal in directions that are prohibited in the linear regime because of momentum conservation, thus giving access to the semiconductor many-body excitations. The typical example is the four-wave mixing spectroscopy.^{1,2} Recently also wave mixing of higher orders started to draw attention.³⁻⁵ A great success in understanding the mechanism of the formation of the nonlinear response in general was achieved in relatively low-field limit using the perturbation theory with respect to the external field.⁶⁻¹² Perturbational description, however, is not suitable for investigating the coherent reconstruction of the spectrum such as in the case of Rabi oscillations. For a qualitative analysis of such phenomena, few-level quantum models have been used.¹³⁻¹⁵ This approach, however, misses the important property of the excitations in semiconductors. These excitations constitute a quantum field rather than a canonical quantum-mechanical system. The system supports infinitely many states, unless of course the spectrum has truly discrete component that is when the excitations are localized such as in quantum dots. Thus a description nonperturbative with respect to the external field must deal with many-body aspects of the dynamics of the system.⁴ In order to treat this problem, a variety of methods based on derivation of respective closed equations of motion was developed.^{2,16-21} Unfortunately, the dynamical equations turn out to be very complex owing to the Coulomb interaction, which is shown to be crucially important at relatively long-time scales.^{22,23} Therefore, one has to resort to numerical calculations when investigating the dynamics of the exciton polarization. The numerical simulations proved to describe successfully the dynamics,^{14,19,24} but they are difficult

to apply for studying the detailed effect of different contributions to the dynamics and its dependence on parameters of the system. As a result, the theory of the semiconductor response suffers from a lack of exact results obtained in controllable approximations which would guide the respective numerical, theoretical, and experimental studies.

At short-time scales, however, one can rely on significant simplification of the dynamics of the system due to negligible phase change during the optical excitation. Effectively, the system follows the external field, which is illustrated best by a simple dynamical model,

$$\dot{P}(t) = -i\omega[P(t)]P(t) + E(t), \quad (1)$$

with $\omega(P)$ being a real positive function. If initially the system is at rest $P(0)=0$, we can present the time dependence $P(t)$ in the form,

$$P(t) = \int_0^t dt' \exp\left[-i \int_{t'}^t dt'' \omega[P(t'')]\right] E(t'). \quad (2)$$

This representation is convenient for comparing the responses of the system with respect to the δ -functional excitation $E(t)=\epsilon\delta(t+0)$ and to the excitation with the piecewise constant amplitude $E(t)=\epsilon/T$, where T is the duration of the forced regime. As follows from Eq. (2), in the case of the δ pulse immediately after the excitation is switched off one has $P(t)=\epsilon$. For the excitation with constant amplitude Eq. (2) yields an estimate maximally different from the δ -pulse case in the form $P(T)=\epsilon(1-e^{-i\omega_m T})/i\omega_m T$, where $\omega_m = \sup_{0 \leq P \leq \epsilon} \omega(P)$. Thus if the excitation pulse is shorter than the time scale determined by ω_m , i.e., if $\omega_m T \ll 1$, then the response with respect to the constant pulse of finite duration differs up to quadratic terms from the δ -pulse response only by the phase factor $e^{-i\omega_m T/2}$.

An immediate application of this consideration to the evolution of the exciton polarization in optically excited semi-

conductors is prevented by the more complex character of the polarization dynamics, namely, by the field induced coupling between the polarization and the charge densities. In order to see the principal differences introduced by this coupling, we consider a model

$$\begin{aligned}\dot{P}(t) &= -i\omega P(t) - iE(t)n(t) - iE(t), \\ \dot{n}(t) &= i\Omega n(t) - iE(t)P(t).\end{aligned}\quad (3)$$

Straightforward substitution of the δ -shaped pulse into this equation obviously fails since now the amplitudes in front of the δ function in the coupling terms are given by the quantities $n(t)$ and $P(t)$, which experience discontinuity at exactly the point of singularity of the δ function. Therefore, we need to consider the case of the external pulse of finite duration. For the pulse with piece-wise constant amplitude $E = \epsilon/T$, as in the previous example, Eq. (3) with the initial conditions $P(0)=0$, $n(0)=0$ is solved for $t \leq T$ by

$$\begin{aligned}P(t) &= -i\frac{2\epsilon}{\Omega T}\sin(\Omega t/2)\left[\sin(\Omega t/2) - i\frac{\omega}{\Omega}\cos(\Omega t/2)\right], \\ n(t) &= -\frac{\epsilon^2}{\Omega^2 T^2}[1 - \cos(\Omega t)],\end{aligned}\quad (4)$$

where $\Omega^2 = \omega^2/4 + E^2$. It is seen that in the limit of the excitation pulses, which are short compared to the typical dynamical time scale $\omega T \ll 1$, one has to distinguish the cases of weak and strong excitations $\omega T/\epsilon \gtrsim 1$ and $\omega T/\epsilon \ll 1$, respectively. In the first case up to the quadratic terms $\Omega^2 T^2$, the response $P(T)$ looks like the one of the system where P and n are decoupled with respect to δ -pulse excitation. This illustrates the relation with the δ -functional approximation popular in studying the nonlinear optical response by using perturbational approach, which corresponds to weak scattering.

In the limit of strong external field when $\omega T \ll 1$ while $\epsilon \gtrsim 1$, the evolution of the system is significantly different. Neglecting the terms quadratic in $\omega T/\epsilon$, we obtain that the evolution of the system is similar to the one we have in the case $\omega=0$ (i.e., Rabi flopping) with the only difference that $P(t)$ acquires a (small) imaginary part. Applying now the similar idea for estimating a phase shift as for the analysis of Eq. (1), we can see that the conclusion about small perturbations vanishing with $T \rightarrow 0$ holds also in the case of P -dependent frequency and renormalized coupling $E \rightarrow E + \eta(|P|, |n|)$.

These results serve as a general background for the analysis of the basic features of the immediate response of a semiconductor excited by short pulses of high intensity. In this regime, while taking exactly into account the many-body effects due to the fermion nature of the electrons and holes, one can neglect the Coulomb interaction and still be able to provide a qualitative description. From this perspective, the roles played by statistics and interactions are clearly different. The statistics impose instantaneous constraints on the dynamics. Interactions, in turn, need time to develop their effect.

Explicitly the possibility to neglect the Coulomb interaction in the semiconductor Bloch equation (SBE) was shown for the two-band model in Ref. 25. In Ref. 23, the numerical investigation of the coupled Maxwell-Bloch equations in multiple quantum well structures has shown that neglecting the Coulomb interaction at a short-time scale a few Rabi flops can be reproduced with good accuracy. The case of very short excitation of very high intensity was studied in Ref. 26 from the perspective of the carrier-wave Rabi flopping.²⁷⁻²⁹ It was demonstrated that even the spectrum of the emitted radiation is approximated remarkably well by the free-carrier model, which corresponds to neglecting the Coulomb interaction in the SBE. It should be noted, however, that this extreme limit should be considered with care because of the failure of the rotating wave approximation.^{27,30}

In the present paper, we extend the consideration of the semiconductor response with respect to the short intensive excitation taking into account multiple hole states in the valence band and the formation of the MWM response. The structure of the paper is as follows. In Sec. II we consider the limiting case when the effect of the internal dynamics can be completely neglected, which corresponds to the cases $\Omega=0$ in the examples considered above. We obtain classes of state related to the spin selection rules and the respective spectrum of the Rabi frequencies. In Sec. III we discuss the effect of the internal dynamics and the Coulomb interaction.

II. RABI OSCILLATIONS

We consider the dynamics of excitation of a semiconductor quantum well during the action of an ultrashort pulse of high intensity, so that the period of the Rabi oscillations is smaller than the typical time scales determined by the internal dynamics. We consider the question of validity of this approximation in Sec. III. We assume that the pulse is tuned to resonance with the exciton levels lying below the band edge and that the envelope of the pulse is constant while the field is on. By means of a canonical transformation, the harmonic time dependence of the excitation field can be excluded (see e.g., Ref. 31) so that in the rotating wave approximation the transformed Hamiltonian becomes time independent. For the description of the semiconductor dynamics, one can use the SBE accounting for degenerate valence bands.^{18,22,32} Having the dynamical equations at hand, one can work out the short-time approximation similarly to how it was done in Sec I. However, in the short-time approximation, it is more convenient to study directly the exciton polarization. In the coherent regime it is defined as $P_\mu = \langle \psi(t) | B_\mu | \psi(t) \rangle$, where $|\psi(t)\rangle$ is the state of the semiconductor, μ denotes the whole set of relevant quantum numbers describing the specific exciton (bound or unbound) state, and B_μ is the exciton annihilation operator defined as $\langle 0 | B_\mu = \langle \mu |$. In terms of the electron and hole operators, the exciton operator B_μ is presented as

$$B_\mu = \int d\mathbf{x} d\mathbf{y} \phi_\mu^*(\mathbf{x}, \mathbf{y}) c_{s_\mu}(\mathbf{x}) v_{\sigma_\mu}(\mathbf{y}), \quad (5)$$

where $\phi_\mu(\mathbf{x}, \mathbf{y})$ is the exciton wave functions, $v_{\sigma_\mu}(\mathbf{y})$ destroys a hole in the valence band with the spin state σ_μ at the

point with the coordinate \mathbf{y} , and $c_{s_\mu}(\mathbf{x})$ destroys an electron in the conduction band with the spin state s_μ at point \mathbf{x} .

Neglecting the Coulomb interaction and the effect of slow internal dynamics determined by small detuning ω_μ means the semiconductor Hamiltonian is approximated by the Hamiltonian of light-matter interaction H_e . In the rotating wave and dipole approximations, the interaction Hamiltonian has the form,^{2,33,34}

$$H_e = \sum_{\mu} [B_{\mu} \epsilon_{\mu}^*(t) + B_{\mu}^{\dagger} \epsilon_{\mu}(t)], \quad (6)$$

where $\epsilon_{\mu}(t)$ is a piece-wise constant function of time, $\epsilon_{\mu}=0$ when the field is off and $\epsilon_{\mu} = \int d\mathbf{x} d\boldsymbol{\mu} \cdot \mathbf{E}(\mathbf{x}) \phi_{\mu}^*(\mathbf{x}, \mathbf{x})$, with $\mathbf{E}(\mathbf{x})$ being the envelope of the external field, when the excitation is on. Here we have introduced \mathbf{d}_{μ} as the respective matrix elements of the dipole moment, which are assumed to be independent on both the electromagnetic field momentum and its angle of incidence.³³ Thus, in this approximation one has $|\psi(t)\rangle = \exp(-iH_e t)|0\rangle$ (throughout the paper, we write formulas using units with $\hbar=1$) and accordingly

$$P_{\mu}(t) = \sum_n \frac{(it)^n}{n!} \langle [H_e, [H_e, \dots [H_e, B_{\mu}] \dots]] \rangle, \quad (7)$$

where $\langle \dots \rangle \equiv \langle 0 | \dots | 0 \rangle$. We calculate this series using the observation that commuting $[B_{\mu}, B_{\mu}^{\dagger}]$ with H_e we arrive at an operator, which can be presented in a form similar to Eq. (5). We employ this observation introducing operators,

$$M^{(n)} = \sum_{\sigma, s} \int d\mathbf{x} d\mathbf{y} [U_{\sigma, s}^{(n)}(\mathbf{x}, \mathbf{y}) v_{\sigma}^{\dagger}(\mathbf{x}) c_s^{\dagger}(\mathbf{y}) + D_{\sigma, s}^{(n)}(\mathbf{x}, \mathbf{y}) c_s(\mathbf{y}) v_{\sigma}(\mathbf{x})], \quad (8)$$

which are related to each other through repetitive commutation with H_e , namely, $M^{(n+1)} = [H_e, [H_e, M^{(n)}]]$. With the ‘‘initial condition’’ $M^{(0)} = B_{\mu}$, the operators $M^{(n)}$ represent the *even* terms in series (7). These terms themselves do not contribute into P_{μ} since $\langle M^{(n)} \rangle = 0$, which in turn follows from $c|0\rangle = 0$ and $\langle 0|c^{\dagger} = 0$. After commuting with H , however, they produce *c*-number terms $A^{(n)} = \langle [H, M^{(n)}] \rangle$ resulting in

$$P_{\mu}(t) = i \sum_{n=0}^{\infty} (-1)^n \frac{t^{2n+1}}{(2n+1)!} A^{(n)}. \quad (9)$$

In order to simplify the expressions, we assume below that the quantum well can be approximated by a two-dimensional (2D) plane. In this case, the exciton states are characterized by the spin states of the electron and hole constituting the exciton, the center of mass momentum in the plane of the well \mathbf{K} , and other quantum numbers n_{μ} so that $|\mu\rangle = |\sigma_{\mu}, s_{\mu}, \mathbf{K}_{\mu}, n_{\mu}\rangle$.

Using the relation between $M^{(n+1)}$ and $M^{(n)}$, we find

$$U_{\sigma, s}^{(n+1)}(\mathbf{x}, \mathbf{y}) = \sum_{\sigma', s'} [U_{\sigma', s'}^{(n)}(\mathbf{x}, \mathbf{y}) \mathcal{E}_{\sigma, s'}^*(\mathbf{x}) \mathcal{E}_{\sigma', s'}^*(\mathbf{x}) + U_{\sigma, s}^{(n)}(\mathbf{x}, \mathbf{y}) \mathcal{E}_{\sigma', s'}(\mathbf{y}) \mathcal{E}_{\sigma', s'}^*(\mathbf{y}) - 2D_{\sigma', s'}^{(n)}(\mathbf{y}, \mathbf{x}) \mathcal{E}_{\sigma, s'}(\mathbf{x}) \mathcal{E}_{\sigma', s'}(\mathbf{y})],$$

$$D_{\sigma, s}^{(n+1)}(\mathbf{x}, \mathbf{y}) = \sum_{\sigma', s'} [D_{\sigma', s'}^{(n)}(\mathbf{x}, \mathbf{y}) \mathcal{E}_{\sigma, s'}^*(\mathbf{x}) \mathcal{E}_{\sigma', s'}(\mathbf{x}) + D_{\sigma, s}^{(n)}(\mathbf{x}, \mathbf{y}) \mathcal{E}_{\sigma', s'}^*(\mathbf{y}) \mathcal{E}_{\sigma', s'}(\mathbf{y}) - 2U_{\sigma', s'}^{(n)}(\mathbf{y}, \mathbf{x}) \mathcal{E}_{\sigma, s'}^*(\mathbf{x}) \mathcal{E}_{\sigma', s'}^*(\mathbf{y})], \quad (10)$$

where $\mathcal{E}_{\sigma, s}(\mathbf{x}) = d_{\sigma, s} E_{s-\sigma}(\mathbf{x})$. In terms of the kernels of the operators $M^{(n)}$, the coefficients $A^{(n)}$ are expressed as $A^{(n)} = \int d\mathbf{x} A^{(n)}(\mathbf{x})$ with

$$A^{(n)}(\mathbf{x}) = \sum_{\sigma, s} [\mathcal{E}_{\sigma, s}^*(\mathbf{x}) U_{\sigma, s}^{(n)}(\mathbf{x}, \mathbf{x}) - \mathcal{E}_{\sigma, s}(\mathbf{x}) D_{\sigma, s}^{(n)}(\mathbf{x}, \mathbf{x})]. \quad (11)$$

Some general results can be obtained directly from Eqs. (10) and (11). First, the polarization of dark excitons (states with helicity 0 and ± 2) is zero. Second, if the excitation pulse is circularly polarized then the summation over the spin indices in Eqs. (10) and (11) reduces to the single terms with the electron and hole spins determined by s_{μ} and σ_{μ} , respectively. If the excitation pulse is linearly polarized, then only the electron spin is fixed to s_{μ} and as a result different electron-hole states get coupled.¹⁸ The summation over the hole spins, as will be explicitly demonstrated below, is limited to such values that meet the condition $\sigma - \sigma_{\mu} \in \{2, 0, -2\}$. For example, if $\sigma_{\mu} = 3/2$ (heavy holes) then σ can take values $3/2$ and $-1/2$ (light hole).

It follows from Eqs. (10) and (11) that $A^{(n+1)}(\mathbf{x}) = \Omega_{\mu}^2(\mathbf{x}) A^{(n)}(\mathbf{x})$, where

$$\Omega_{\mu}^2(\mathbf{x}) = 4 \sum_{\sigma = \sigma_{\mu}, \bar{\sigma}_{\mu}} |\mathcal{E}_{\sigma, s_{\mu}}(\mathbf{x})|^2, \quad (12)$$

with $\bar{\sigma}_{\mu}$ being the second value satisfying the spin selection rule as discussed above. Applying these results to Eq. (9) we find the general representation, which is valid for an excitation pulse with an arbitrary spatial profile in the plane of the quantum well,

$$P_{\mu}(t) = -i \int d\mathbf{x} \frac{1}{\Omega_{\mu}(\mathbf{x})} \mathcal{E}_{\mu}(\mathbf{x}) \phi_{\mu}^*(\mathbf{x}, \mathbf{x}) \sin[\Omega_{\mu}(\mathbf{x}) t]. \quad (13)$$

This result can be generalized for the more general case, when the envelope amplitude of the external field is not a piece-wise constant but can be presented as $\mathbf{E}(\mathbf{x}, t) = f(t)\mathbf{E}(\mathbf{x})$. We note that the Hamiltonians $H_e'(t) = f(t)H_e$ taken at different instants t_1 and t_2 commute with each other. Therefore, the line of arguments used above can be repeated with the substitution,

$$t \rightarrow \int_0^t dt' f(t'). \quad (14)$$

It can be easily seen that the expression for the exciton polarization $P_{\mu}(t)$ remains essentially the same,

$$P_{\mu}(t) = -i \int d\mathbf{x} \frac{1}{\Omega_{\mu}(\mathbf{x}, t)} \mathcal{E}_{\mu}(\mathbf{x}, t) \phi_{\mu}^*(\mathbf{x}, \mathbf{x}) \sin[\varpi_{\mu}(\mathbf{x}, t)], \quad (15)$$

where $\mathcal{E}_{\mu}(\mathbf{x}, t)$ and $\Omega_{\mu}(\mathbf{x}, t)$ are defined by the same expressions as above but with the time-dependent envelope func-

tion of the external field, i.e., they differ from $\mathcal{E}_\mu(\mathbf{x})$ and $\Omega_\mu(\mathbf{x})$ by the factor $f(t)$. The phases

$$\varpi_\mu(\mathbf{x}, t) = \int_0^t dt' \Omega_\mu(\mathbf{x}, t'), \quad (16)$$

have the meaning of the pulse areas. Since such generalization does not add to the physical picture but complicates the discussion of the time dependence, in what follows we will consider only the case of piece-wise constant amplitude of the external excitation.

A. Single-pulse response

First we briefly discuss the case when the excitation is a single plane wave $\mathbf{E}(\mathbf{x}) = (E_+ \hat{\mathbf{e}}_+ + E_- \hat{\mathbf{e}}_-) e^{i\mathbf{K}_0 \cdot \mathbf{x}}$. In this case, $\Omega_\mu(\mathbf{x})$ does not depend on the coordinate and the integration in Eq. (13) can be easily performed taking into account that $\phi_\mu^*(\mathbf{x}, \mathbf{y}) = e^{-i\mathbf{K}_\mu \cdot \mathbf{R}_\mu} \phi_\mu^*(\mathbf{x} - \mathbf{y})$, where \mathbf{R}_μ is the coordinate of the center of mass of the exciton and $\phi_\mu^*(\mathbf{x} - \mathbf{y})$ is its relative wave function. For example, for $(h+, 1s)$ exciton polarization, we obtain

$$P_{h+, \mathbf{K}}(t) = -i \delta(\mathbf{K} - \mathbf{K}_0) d_{h+} E_+ \phi_{h+}^*(0) \frac{1}{\Omega_{h+}} \sin(\Omega_{h+} t), \quad (17)$$

where $\phi_{h+}^*(0)$ is the value of the exciton wave function at the origin, which in the 2D approximation is $\phi_{h+}^*(0) = r_{h+}^{-1} \sqrt{2/\pi}$, with r_{h+} being the respective exciton Bohr radius and

$$\Omega_{h+} = 2\sqrt{|d_{h+} E_+|^2 + |d_{l-} E_-|^2}. \quad (18)$$

Equation (17) presents the Rabi oscillations of the exciton polarization. It is interesting to note that this result explicitly shows the statistical origin of the Rabi oscillations. Following the same line of arguments, one can show that the oscillations would be absent and the exciton polarization would increase monotonously with time if the exciton operators had obeyed bosonic commutation relations or if the electrons and holes were bosons. Indeed, let, for example, the exciton operators be bosonic then all terms in Eq. (7) with $n > 1$ turn to zero leaving $P_\mu \propto t$.

Because of the coupling between different exciton states, the evolution of the exciton polarization is characterized by two Rabi frequencies $\Omega_{h+} = \Omega_{l-}$ and $\Omega_{h-} = \Omega_{l+} = 2\sqrt{|d_{h-} E_-|^2 + |d_{l+} E_+|^2}$. The ratio between these frequencies is shown in Fig. 1 as a function of E_+/E_- , where (for calculations) we have taken³⁵ $d_h/d_l = \sqrt{3}$.

The effect of sharing the Rabi frequency leads to the same time dependence of the heavy- and light-hole contributions into the signal with fixed helicity if the excitation is linearly polarized. If, however, the external field has elliptic polarization these contributions oscillate with different frequencies leading to beatings and to a nontrivial time dependence of the polarization state of the signal.

B. Operator equations of motion

Before we proceed, we would like to discuss in details the spin selection rules noticed above from the perspective of the

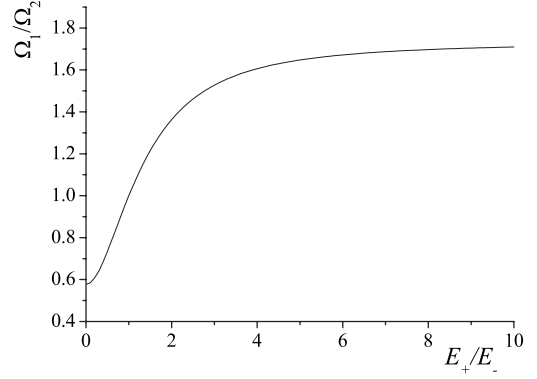


FIG. 1. The ratio of two Rabi frequencies Ω_{h+}/Ω_{h-} as a function of ellipticity of the excitation pulse E_+/E_- . The ratio varies from $|d_l/d_h|$ to $|d_h/d_l|$. For calculations, the ratio of heavy- and light-hole dipole moments is taken $d_h/d_l = \sqrt{3}$.

operator equations of motion. For this we consider an operator of the form

$$\begin{aligned} \hat{F}(t; \mathbf{x}, \mathbf{y}) = & f_{\sigma_1, \sigma_2}(t) v_{\sigma_1}^\dagger(\mathbf{x}) v_{\sigma_2}(\mathbf{y}) + g_{s_1, s_2}(t) c_{s_1}^\dagger(\mathbf{x}) c_{s_2}(\mathbf{y}) \\ & + \pi_{\sigma, s}^{(1)}(t) v_{\sigma}^\dagger(\mathbf{x}) c_s^\dagger(\mathbf{y}) + \pi_{\sigma, s}^{(2)}(t) c_s(\mathbf{y}) v_{\sigma}(\mathbf{x}) \\ & + A(t) \delta(\mathbf{x} - \mathbf{y}), \end{aligned} \quad (19)$$

where the sum is taken over all spin indices. The amplitudes in this expression are chosen in such a way that $\hat{F}(t; \mathbf{x}, \mathbf{y})$ is the Heisenberg representation of the operator $\hat{F}(0; \mathbf{x}, \mathbf{y})$ defined by the light-matter Hamiltonian,

$$\hat{F}(t; \mathbf{x}, \mathbf{y}) = e^{iH_e t} \hat{F}(0; \mathbf{x}, \mathbf{y}) e^{-iH_e t}. \quad (20)$$

The last term in Eq. (19) determines the average value $\langle \hat{F}(t; \mathbf{x}, \mathbf{y}) \rangle$ and for the case of the exciton polarization was considered above. For the purpose of our discussion, it suffices to consider the dynamical equations for the time-dependent amplitudes $g_{\sigma_1, \sigma_2}(t)$ and so on for the case of $\mathbf{x} = \mathbf{y}$ and spatially independent external field $\mathcal{E}_{\sigma, s}$. From the Heisenberg equation of motion $-i\dot{\hat{F}} = [\hat{H}_e, \hat{F}]$, we find (see Appendix A)

$$\begin{aligned} \dot{f}_{s_1, s_2} &= i\mathcal{E}_{\sigma', s_1} \pi_{\sigma', s_2}^{(2)} - i\pi_{\sigma', s_1}^{(1)} \mathcal{E}_{\sigma', s_2}^*, \\ \dot{g}_{\sigma_1, \sigma_2} &= i\mathcal{E}_{\sigma_1, s'} \pi_{\sigma_2, s'}^{(2)} - i\pi_{\sigma_1, s'}^{(1)} \mathcal{E}_{\sigma_2, s'}^*, \\ \dot{\pi}_{\sigma, s}^{(1)} &= -i\mathcal{E}_{\sigma, s'} f_{s, s'} - i\mathcal{E}_{\sigma', s} g_{\sigma, \sigma'}, \\ \dot{\pi}_{\sigma, s}^{(2)} &= i\mathcal{E}_{\sigma, s'}^* f_{s', s} + i\mathcal{E}_{\sigma', s}^* g_{\sigma', \sigma}, \end{aligned} \quad (21)$$

where the summation over the dashed spin variables is implied. With the respective initial conditions, these equations define the Heisenberg representation of the operator $\hat{F}(0; \mathbf{x}, \mathbf{y})$. For example, if all amplitudes but $\pi_{\sigma, s}^{(1)}$ are initially zero, then this would be the representation of the *interband* polarization creation operator and so on. It should be noted that no restrictions on the amplitudes being “bright” or “dark” are imposed in Eq. (21). Thus this is a system with

respect to 36 unknowns and, counting degeneracy, has as many characteristic frequencies. The actual number of different frequencies, however, turns out to be much smaller.

Taking the derivative of the last pair of equations with respect to time, we obtain the closed system

$$\begin{aligned}\ddot{\pi}_{\sigma,s}^{(1)} &= -\mathcal{E}_{\sigma,s'}\mathcal{E}_{\sigma',s}^*\pi_{\sigma',s}^{(1)} - \pi_{\sigma,s}^{(1)}\mathcal{E}_{\sigma',s'}\mathcal{E}_{\sigma',s} + 2\mathcal{E}_{\sigma,s'}\pi_{\sigma',s}^{(2)}\mathcal{E}_{\sigma',s}, \\ \ddot{\pi}_{\sigma,s}^{(2)} &= -\mathcal{E}_{\sigma,s'}^*\mathcal{E}_{\sigma',s'}\pi_{\sigma',s}^{(2)} - \pi_{\sigma,s}^{(2)}\mathcal{E}_{\sigma',s'}\mathcal{E}_{\sigma',s} + 2\mathcal{E}_{\sigma,s'}^*\pi_{\sigma',s}^{(1)}\mathcal{E}_{\sigma',s},\end{aligned}\quad (22)$$

where again the summation over dashed spin indices is performed. Taking the complex conjugation of these equations, we obtain the same system but with respect to $[\pi^{(2)*}, \pi^{(1)*}]$. Because of this symmetry the solutions of Eq. (22) are divided into two classes $\pi^{(2)} = \pi^{(1)*}$ and $\pi^{(2)} = -\pi^{(1)*}$, which correspond to *A* and *B* irreducible representations of the group Z_2 , respectively. Additional simplifications come from the structure of the coupling induced by the external field. Having in mind the successive application for the dynamics determined by the SBE, we consider the additional symmetries on a general ground. Due to the electric dipole selection rules, only four elements of $\mathcal{E}_{\sigma,s}$ are nonzero,

$$\mathcal{E}_{\sigma,s} = \begin{pmatrix} \mathcal{E}_{h-} & 0 \\ 0 & \mathcal{E}_{l-} \\ \mathcal{E}_{l+} & 0 \\ 0 & \mathcal{E}_{h+} \end{pmatrix}, \quad (23)$$

where the columns are enumerated by the electron-spin projections $1/2, -1/2$, and rows denote different hole spins $3/2, \dots, -3/2$. We denote by $\mathcal{V}_\mathcal{E}$ the vector space consisting of the elements of form (23) but without any relation between the nonzero elements, that is, the four-dimensional subspace of the interband polarizations. For two elements u and $v \in \mathcal{V}_\mathcal{E}$, we consider $f = v^T u$ where the composition is the usual matrix multiplication of two (rectangular) matrices,

$$f_{s_1, s_2} = \sum_{\sigma'} v_{\sigma', s_1} u_{\sigma', s_2}. \quad (24)$$

The space spanned by such elements will be called \mathcal{V}_e . This is a 2D subspace of the four-dimensional space of the electron densities. Using Eq. (23) in (24) one can see that in the basis of the electron-hole spins, \mathcal{V}_e consists of diagonal 2×2 matrices. Finally, for the elements u and v we consider $g = v u^T$, which are the matrices with the matrix elements,

$$g_{\sigma_1, \sigma_2} = \sum_{s'} v_{\sigma, s'} u_{\sigma_2, s'}. \quad (25)$$

We will call the respective linear vector space \mathcal{V}_h , which (as can be easily checked) is eight-dimensional subspace of the 16-dimensional space of the hole-hole correlation functions.

The elements of \mathcal{V}_e and \mathcal{V}_h can be considered as operators acting on the electron and the hole spin variables, respectively. The important properties of these operators is that their action is reducible, namely, \mathcal{V}_e leaves the spin state intact and \mathcal{V}_h mixes only states within the groups $\{3/2, -1/2\}$ and $\{-3/2, 1/2\}$ but not from the different groups. As

a result when \mathcal{V}_e and \mathcal{V}_h act on $\mathcal{V}_\mathcal{E}$ (from the left and from the right, respectively), they leave $\mathcal{V}_\mathcal{E}$ intact.

Using these results, we obtain the modes of system (22). These modes are naturally classified by the invariant spin classes, which do not mix during the evolution, and by the irreducible representations of the group Z_2 . Inspecting Eq. (22) one can see that the spin classes in the spin space of the electron-hole polarizations are $C_1 = \{(-3/2, -1/2), (1/2, -1/2)\}$ or $\{h+, l-\}$, $C_2 = \{(3/2, 1/2), (-1/2, 1/2)\}$ and $C_3 = \{(3/2, -1/2), (1/2, 1/2), (-1/2, -1/2), (-3/2, 1/2)\}$. The first two classes correspond to the bright exciton operators; the last class contains the dark excitons.

1. Bright exciton classes $C_{1,2}$

In each class both states have the same projection of the electron spin, $s=1/2$ for C_1 and $s=-1/2$ for C_2 , while the hole spin can take two values. If we need to distinguish these values we will denote them σ and $\bar{\sigma}$. In order to find the modes one can present $\pi_{\sigma,s}^{(1)}(t) = \pi_{\sigma,s}^{(1)} \sin(\omega t)$ in Eq. (22). Studying the resulting system of homogeneous equations, one finds the frequencies and (non-normalized) eigenvectors of the modes,

$$\omega^2 = 4\beta_s^2, \quad \pi_{\sigma,s}^{(1)} = \mathcal{E}_{\sigma,s}, \quad (B)$$

$$\omega^2 = \beta_s^2, \quad \pi_{\sigma,s}^{(1)} = \frac{1}{\mathcal{E}_{\sigma,s}}, \quad \pi_{\bar{\sigma},s}^{(1)} = -\frac{1}{\mathcal{E}_{\bar{\sigma},s}}, \quad (A, B)$$

$$\omega^2 = 0, \quad \pi_{\sigma,s}^{(1)} = \mathcal{E}_{\sigma,s}, \quad (A), \quad (26)$$

where $\beta_s^2 = |\mathcal{E}_{\sigma,s}|^2 + |\mathcal{E}_{\bar{\sigma},s}|^2$ [$4\beta_s^2$ coincides with the frequency defined in Eq. (12)] and *A* and *B* denote the even and odd irreducible representations, respectively. Using these solutions in the first two equations of Eq. (21), we find the electron and hole densities coupled to the respective interband polarization operators. One can immediately see that these densities belong to \mathcal{V}_e and \mathcal{V}_h , respectively.

As immediately follows from Eqs. (11) and (A2), only *B* solutions can contribute into the average value of the exciton polarization since for the symmetric solutions both terms under the integral in Eq. (A2) cancel each other. Moreover, for the *B* solutions with the frequencies $\omega^2 = \beta_s^2$, the sum $\pi_{\sigma,s}^{(1)}\mathcal{E}_{\sigma,s} + \pi_{\bar{\sigma},s}^{(1)}\mathcal{E}_{\bar{\sigma},s}$ vanishes, which leaves the only solution with $\omega^2 = 4\beta_s^2$ in accordance with Eq. (13).

2. Dark exciton class C_3

The dark exciton operators are specified by the amplitudes in the complement to $\mathcal{V}_\mathcal{E}$, i.e., in the four-dimensional subspace spanned by such elements that have zeros instead of nonzero elements in Eq. (23) and vice versa. The states are naturally enumerated by the hole spin state σ and for each amplitude $\pi_{\sigma,s}$ there is a correspondent component of the excitation field obtained by inversion of the electron spin $\mathcal{E}_{\sigma,\bar{s}}$ with $\bar{s} = -s$. The frequencies and nonzero components of different modes are found to be

$$\omega^2 = \beta_{1/2}^2, \quad \pi_{3/2,-1/2}^{(1)} = -\frac{1}{\mathcal{E}_{3/2,1/2}},$$

$$\pi_{-1/2,-1/2}^{(1)} = \frac{1}{\mathcal{E}_{-1/2,1/2}}, \quad (A, B)$$

$$\omega^2 = \beta_{-1/2}^2, \quad \pi_{1/2,1/2}^{(1)} = -\frac{1}{\mathcal{E}_{1/2,-1/2}},$$

$$\pi_{-3/2,1/2}^{(1)} = \frac{1}{\mathcal{E}_{-3/2,-1/2}}, \quad (A, B)$$

$$\omega^2 = (\beta_{1/2} \pm \beta_{-1/2})^2, \quad \pi_{3/2,-1/2}^{(1)} = \pi_{-1/2,-1/2}^{(1)}$$

$$= \mp \beta_{1/2}, \quad \pi_{-3/2,1/2}^{(1)} = \pi_{1/2,1/2}^{(1)} = \mp \beta_{-1/2}, \quad (A),$$

$$\omega^2 = (\beta_{1/2} \pm \beta_{-1/2})^2, \quad \pi_{3/2,-1/2}^{(1)} = \pi_{-1/2,-1/2}^{(1)}$$

$$= \pm \beta_{1/2}, \quad \pi_{-3/2,1/2}^{(1)} = \pi_{1/2,1/2}^{(1)} = \pm \beta_{-1/2}. \quad (B)$$

(27)

Since for each $\pi_{\sigma,s}$ in this class the respective components of the external field are zeros, i.e., $\mathcal{E}_{\sigma,s}=0$, the dark exciton amplitudes [according to Eq. (A2)] do not contribute to $\dot{A}(t)$. Thus the average value of the dark exciton polarization equals to its initial value $\hat{F}_{\sigma,s}(t) = \hat{F}_{\sigma,s}(0)$, where $\hat{F}_{\sigma,s}(0) \propto v_{\sigma}^{\dagger} c_s^{\dagger}$ with σ and s from C_3 . If the initial state of the system is vacuum then $\langle \hat{F}_{\sigma,s}(t) \rangle = 0$ —confirming the result about absent polarization of the dark excitons.

3. Operator dynamics

The frequencies found $\omega^2 = 4\beta_s^2, \beta_s^2, (\beta_s \pm \beta_s)^2$ define all the frequencies introduced into the dynamics due to the interaction with the external field as defined by Eq. (21). The remaining eight frequencies can be shown to be zero. Four solutions (two for each class C_1 and C_2) corresponding to the zero frequency are presented in Eq. (26) and the remaining four can be found from the first two equations of Eq. (21). Since Eq. (21) has the form of a Schrödinger equation with a Hermitian Hamiltonian, the solutions corresponding to $\omega = 0$ are not secular solutions but rather invariants, that is they correspond to operators, which are invariant with respect to action of the external field. For example [as follows from Eq. (26)], for the exciton polarization such operators are $\sum_{\sigma,s} \mathcal{E}_{\sigma,s} v_{\sigma}^{\dagger} c_s^{\dagger} + \text{H.c.}$, where the spin summation is restricted to a particular class C_1 or C_2 .

In the short-time limit only frequencies $4\beta_s^2$ contribute to the Rabi oscillations of the exciton polarization. However, as will be demonstrated later, the slow dynamics also lead to admixture of other frequencies.

C. Multiwave mixing response

A more complex situation arises when the excitation pulse consists of two plane waves with noncollinear wave vectors $\mathbf{E}(\mathbf{K}) = \mathbf{E}^{(1)} \delta(\mathbf{K} - \mathbf{K}_1) + \mathbf{E}^{(2)} \delta(\mathbf{K} - \mathbf{K}_2)$. This corresponds to the standard problem of excitation by two pulses with zero delay between them. Factoring out the exponential factor corre-

sponding to the first pulse, we can rewrite Eq. (13) as

$$\int d\mathbf{x} e^{i\mathbf{k}\cdot\mathbf{x}} F(\mathbf{x}), \quad (28)$$

where $F(\mathbf{x})$ is a periodic function $F[\mathbf{x} + \Delta\mathbf{K}/(2\pi\Delta K^2)] = F(\mathbf{x})$ with $\Delta\mathbf{K} = \mathbf{K}_2 - \mathbf{K}_1$. Thus, integration over \mathbf{x} yields the exciton polarization in the form,

$$P_{\mu}(t) = \sum_{m=-\infty}^{\infty} P_{\mu}^{(m)}(t) \delta(\mathbf{K}_{\mu} - \mathbf{K}_1 - m\Delta\mathbf{K}), \quad (29)$$

where $P_{\mu}^{(m)}(t)$ are the amplitudes of multiwave mixing (MWM) polarizations. The amplitudes $P_{\mu}^{(0)}(t)$ and $P_{\mu}^{(1)}(t)$ correspond to the polarization along the directions of linear response \mathbf{K}_1 and \mathbf{K}_2 , respectively. For $m = -1, 2$ one has four-wave mixing polarization, $m = -2, 3$ correspond to six-wave mixing, and so on. Performing the Fourier transform of the periodic factor $F(\mathbf{x})$, we derive the integral representation for the amplitudes of MWM polarization,

$$P_{\mu}^{(m)}(t) = -\frac{i\phi_{\mu}^{*}(0)}{2\pi} \int_0^{2\pi} d\theta e^{-im\theta} \frac{\mathcal{E}_{\mu}^{(1)} + \mathcal{E}_{\mu}^{(2)} e^{i\theta}}{|\Omega_{\mu}^{(1)} + \Omega_{\mu}^{(2)} e^{i\theta}|}$$

$$\times \sin(|\Omega_{\mu}^{(1)} + \Omega_{\mu}^{(2)} e^{i\theta}|t), \quad (30)$$

where $\mathcal{E}_{\mu}^{(i)} = d_{\mu} E_{\mu}^{(i)}$ and the frequencies $\Omega_{\mu}^{(1,2)}$ are defined as the amplitudes of $\Omega_{\mu}(\mathbf{x})$ corresponding to different pulses $\Omega_{\mu}^{(i)} = 2\sqrt{\sum_{\sigma} |\mathcal{E}_{\sigma,s}^{(i)}|^2}$.

In Fig. 2, we show the numerical evaluations of Eq. (30) for different ratios between the amplitudes of the plane waves constituting the excitation along one of the forward and four-wave mixing directions. The initial stage of the dynamics for $t \rightarrow 0$ can be obtained directly from series (9) in the form (for $m > 0$),

$$P_{\mu}^{(m)}(t) = i \frac{(-1)^m \phi_{\mu}^{*}(0) \mathcal{E}_{\mu}^{(2)}}{(2m-1)! \eta_{\mu}} (\eta_{\mu} t)^{2m-1}, \quad (31)$$

where $\eta_{\mu} = \sqrt{\Omega_{\mu}^{(1)} \Omega_{\mu}^{(2)}}$.

The evolution of the exciton polarization in the opposite limit $t \rightarrow \infty$ is found from integral representation (30) using the stationary phase method,

$$P_{\mu}^{(m)}(t) = -i \frac{\phi_{\mu}^{*}(0)}{\sqrt{2\pi\eta_{\mu}^2 t}} \left\{ \text{sign}(\mathcal{E}_{\mu}^{(1)} - \mathcal{E}_{\mu}^{(2)}) (-1)^m \frac{|\mathcal{E}_{\mu}^{(1)} - \mathcal{E}_{\mu}^{(2)}|}{\sqrt{|\Omega_{\mu}^{(1)} - \Omega_{\mu}^{(2)}|}} \right.$$

$$\times \sin \left[|\Omega_{\mu}^{(1)} - \Omega_{\mu}^{(2)}| t + \frac{\pi}{4} \right]$$

$$\left. + \frac{\mathcal{E}_{\mu}^{(1)} + \mathcal{E}_{\mu}^{(2)}}{\sqrt{|\Omega_{\mu}^{(1)} + \Omega_{\mu}^{(2)}|}} \sin \left[(\Omega_{\mu}^{(1)} + \Omega_{\mu}^{(2)}) t - \frac{\pi}{4} \right] \right\}. \quad (32)$$

It shows that in this limit, the $P_{\mu}^{(m)}(t)$ demonstrates power decay $\propto t^{-1/2}$ and is essentially a superposition of two harmonics with frequencies $|\Omega_{\mu}^{(1)} \pm \Omega_{\mu}^{(2)}|$. Of course, the limit $t \rightarrow \infty$ should not be taken too literally, since in this limit the main assumption $\omega_{\mu} t \ll 1$ is no longer valid and the effect of desynchronization of the MWM polarization of high orders (see below) becomes important. However, if $\Omega_{\mu}^{(1)}$ and $\Omega_{\mu}^{(2)}$ are not too close then after just a few oscillations the depen-

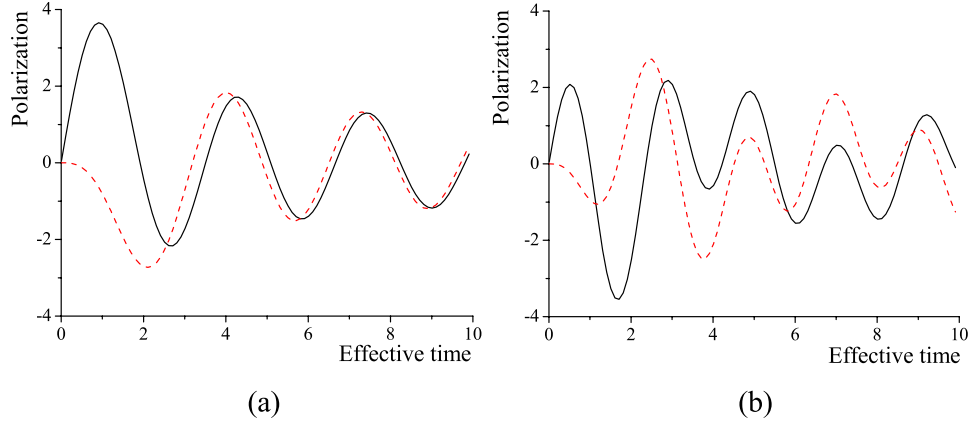


FIG. 2. (Color online) The exciton polarization in the case when the excitation consists of two plane waves with noncollinear wave vectors. The solid and dashed lines show the response in the forward ($m=1$) and the four-wave mixing ($m=2$) directions for (a) $\Omega_\mu^{(1)}/\Omega_\mu^{(2)}=1$ and (b) $\Omega_\mu^{(1)}/\Omega_\mu^{(2)}=2$. The effective time is defined as $\tilde{t}=\Omega_\mu^{(2)}t$.

dence $P_\mu^{(m)}(t)$ is satisfactorily approximated by its asymptotic form. For example, the complex behavior of the exciton polarization shown in Fig. 2(b) is a result of existence of two harmonics with the frequencies $|\Omega_\mu^{(1)} \pm \Omega_\mu^{(2)}|$, which exist if $\Omega_\mu^{(1)} \neq \Omega_\mu^{(2)}$.

If $\Omega_\mu^{(1)} = \Omega_\mu^{(2)} = \Omega_\mu$ then the slow harmonic vanishes. Indeed, in this case [as follows from Eq. (30)], MWM polarizations can be presented in the form,

$$P_\mu^{(m)}(t) = i(-1)^m \phi_\mu^*(0) \left\{ \frac{\mathcal{E}_\mu^{(2)}}{\Omega_\mu} J_{2m-1}(2\Omega_\mu t) - [\mathcal{E}_\mu^{(1)} - \mathcal{E}_\mu^{(2)}] \int_0^t dt' J_{2m}(2\Omega_\mu t') \right\}, \quad (33)$$

where $J_m(t)$ are the Bessel functions of the first kind. This solution asymptotically oscillates with the frequency $2\Omega_\mu$, which is consistent with the simple picture of the Rabi frequency renormalized due to doubling the external field.

The interesting difference between the cases of the single-pulse and two-pulses excitations is that the latter asymptotically decays in time $\propto t^{-1/2}$. It can be understood taking into account the dynamical origin of the formation of the MWM polarization. As follows from Eq. (30), the MWM polarizations $P_\mu^{(m)}(t)$ satisfy the equation of motion of a classical tight-binding model,

$$-\ddot{P}_\mu^{(m)}(t) = \tilde{\Omega}_\mu^2 P_\mu^{(m)}(t) + \eta_\mu^2 P_\mu^{(m+1)}(t) + \eta_\mu^2 P_\mu^{(m-1)}(t), \quad (34)$$

where $\tilde{\Omega}_\mu^2 = \Omega_\mu^{(1)2} + \Omega_\mu^{(2)2}$. Equation (34) is supplemented by the initial conditions $P_\mu^{(m)}(0) \equiv 0$ for all m and $\dot{P}_\mu^{(m)}(0) = 0$ for all m except $m=0, 1$ for which one has $\dot{P}_\mu^{(0)}(0) = -i\mathcal{E}^{(1)}\phi_\mu^*(0)$ and $\dot{P}_\mu^{(1)}(0) = -i\mathcal{E}^{(2)}\phi_\mu^*(0)$. This representation allows one to estimate the effect of the dependence of the exciton energy on the in-plane momentum. It leads to the modification of the ‘‘eigenfrequency’’ $\tilde{\Omega}_\mu^2$ by the term $\sim (m\Delta K)^2/2m_\mu$. Even if $(\Delta K)^2/2m_\mu \ll \tilde{\Omega}_\mu^2$, this contribution becomes essential for MWM exciton polarizations of significantly high orders. However, for the most important case of MWM polarizations of low orders on the time scale of few Rabi flops, the effect

of the correction due to the exciton dispersion can be neglected.

It follows from representation (34) that initially the excitation is localized at the sites $m=0, 1$ (the directions of linear response). With time the energy spreads along the chain thanks to the coupling $\propto \eta_\mu^2$ between different sites. In terms of the MWM polarization, the dynamics of this spreading corresponds to excitation MWM polarizations of higher and higher orders. The dynamical model described by Eq. (34) has the first integral,

$$\sum_m \{ |\dot{P}_\mu^{(m)}(t)|^2 + \tilde{\Omega}_\mu^2 |P_\mu^{(m)}(t)|^2 + \eta_\mu^2 P_\mu^{(m)*}(t) [P_\mu^{(m-1)}(t) + P_\mu^{(m+1)}(t)] \} = |\phi_\mu(0)|^2 (|\mathcal{E}^{(1)}|^2 + |\mathcal{E}^{(2)}|^2). \quad (35)$$

From this perspective, it is clear that the decay of the Rabi oscillations is the consequence of spreading the excitation among MWM polarizations of different orders.

III. EFFECT OF INTERNAL DYNAMICS AND THE COULOMB INTERACTION

In Sec. II we have studied in details the dynamics of the exciton polarization in the short-time limit, when the main contribution into dynamics comes from the interaction with the external field. Here, we consider the effect of the internal semiconductor dynamics and study how the results of Sec. II appear in the framework of a more general description. We approach this problem describing the dynamics of the interband polarizations $p_{\sigma,s}(\mathbf{x}_1, \mathbf{x}_2) = \langle c_s(\mathbf{x}_2) v_\sigma(\mathbf{x}_1) \rangle$, the electron-electron $e_{s_1, s_2}(\mathbf{x}_1, \mathbf{x}_2) = \langle c_{s_1}^\dagger(\mathbf{x}_1) c_{s_2}(\mathbf{x}_2) \rangle$ and the hole-hole $h_{\sigma_1, \sigma_2}(\mathbf{x}_1, \mathbf{x}_2) = \langle v_{\sigma_1}^\dagger(\mathbf{x}_1) v_{\sigma_2}(\mathbf{x}_2) \rangle$ correlation functions by the semiconductor Bloch equation (see Appendix B).

First we notice that the most significant contribution of the full dynamics would be the production of the response, which was absent in the approximation used in Sec. II. This would be generating the interband polarization corresponding to dark states (class C_3). It can be proven, however, that the dynamics described by Eq. (B5) does not support excitation of the dark states. Indeed, integrating both sides of the

dynamical equations with respect to time in the interval $(0, \Delta t)$ with $\Delta t \rightarrow 0$, one can see that initially the spin state of the interband polarization belongs to \mathcal{V}_e while the electron and hole correlation functions are zero and, hence, belong to \mathcal{V}_e and \mathcal{V}_h , respectively. Next, considering the integral over the interval $(t, t+\Delta t)$ one can see that if at instant t the state of the system, i.e., p , h , and e belongs to \mathcal{V}_e , \mathcal{V}_h , and \mathcal{V}_e , respectively; then so does the state at $t+\Delta t$. It should be noted, thereby, that in quantum wells the valence-band mixing does not lead to a violation of invariance of the spin classes, since the Hamiltonian describing the mixing is an element of \mathcal{V}_h . Thus, if initially the system is in the ground state then the interband polarization corresponding only to the bright states will be produced at least as long as the SBE and the approximation of angular independence of the dipole moment hold. In order to describe the dynamical coupling between the bright and dark excitons, one has to study a generalization of the SBE that contains higher-order correlation functions besides single-particle ones that are considered here. For example, the pair of two bright excitons can transform into the pair of dark excitons through carrier exchange (see, e.g., Ref. 36). However, the SBE, truncated at the single-particle level does not take carrier exchange between two excitons into account, which is a genuine two-particle correlation. In order to account for this carrier exchange, one would need to truncate the SBE at higher order, i.e., take higher-order correlation functions into account. However, the analysis of this generalization of the SBE is beyond the scope of this paper.

As the next step, we consider the limit of very short-time response such that following the analysis of the simple model (3) the contribution of the kinetic-energy terms can be neglected, while other terms including nonlinear are kept. The solution found in Sec. II [Eq. (13)] suggests the interband polarization in the form $p(\mathbf{x}, \mathbf{y}) \propto \delta(\mathbf{x} - \mathbf{y})$. Substituting this ansatz into Eq. (B5) with neglected kinetic-energy terms and introducing a cutoff of the Coulomb potential, such that $V(0) = V_0 < \infty$, one arrives at the system of equations independent of V_0 , which is structurally similar to Eq. (A2) and which yields the same result for the exciton polarization as Eq. (13). The cancelation of the terms proportional to the Coulomb potential is particularly evident from the representation of the SBE in form (B3).

This consideration shows that the most significant correction to the approximate solution results from the kinetic-energy terms rather than from the Coulomb interaction—as one might expect considering that the Coulomb interaction is responsible for the nonlinear terms in the SBE.

We would like to emphasize at this point that the quantity of our main interest is the exciton polarization P_μ since these are the excitons that constitute true single-particle semiconductor states. As follows from Eq. (5), P_μ is related to the interband polarization $p_{\sigma,s}$ through

$$P_\mu = \int d\mathbf{x} d\mathbf{y} \phi_\mu^*(\mathbf{x}, \mathbf{y}) p_{\sigma_\mu, s_\mu}(\mathbf{y}, \mathbf{x}). \quad (36)$$

For bound exciton states, this naturally introduces a characteristic spatial scale; the exciton Bohr radius r_B . This circum-

stance, together with the assumption that the typical spatial variation of the external excitation in the plane of the quantum well is small compared to the Bohr radius $K_{1,2} r_B \ll 1$, allows one to simplify the SBE using the parametric approximation. For this we introduce $\mathbf{R} = (\mathbf{x} + \mathbf{y})/2$ and $\mathbf{r} = \mathbf{x} - \mathbf{y}$. Next we notice that (in the case of single-pulse or two-pulse excitations) the solutions of the SBE are invariant with respect to either infinitesimal or finite (by $\Delta \mathbf{K}/(2\pi\Delta K^2)$) translations in \mathbf{R} plane. Having in mind the consecutive convolution of the solutions with the exciton wave function, we can neglect the terms $\propto \Delta K r_B$ and leave only the dependence on \mathbf{R} as a parameter.^{2,18,32} Thus, assuming for simplicity that $\mathbf{K}_1 = 0$, we approximate Eq. (B5) by

$$\begin{aligned} i\dot{p}_{\sigma,s}(\mathbf{r}, \mathbf{R}) &= \tilde{K}_{\sigma,s}[p] + \mathcal{E}_{\sigma,s}(\mathbf{R})\delta(\mathbf{r}) \\ &\quad - \int d\mathbf{r}' [\tilde{\mathcal{E}}_{\sigma',s}(\mathbf{r}', \mathbf{R}) h_{\sigma',\sigma}(\mathbf{r}' - \mathbf{r}, \mathbf{R}) \\ &\quad - \tilde{\mathcal{E}}_{\sigma,s'}(\mathbf{r}', \mathbf{R}) e_{s',s}(\mathbf{r}' + \mathbf{r}, \mathbf{R})], \\ -i\dot{e}_{s_1,s_2}(\mathbf{r}, \mathbf{R}) &= \hat{K}_{s_1,s_2}[e] + \int d\mathbf{r}' [\tilde{\mathcal{E}}_{\sigma',s_1}^*(\mathbf{r}', \mathbf{R}) p_{\sigma',s_2}(\mathbf{r}' + \mathbf{r}, \mathbf{R}) \\ &\quad - \tilde{\mathcal{E}}_{\sigma',s_2}(\mathbf{r}', \mathbf{R}) p_{\sigma',s_1}^*(\mathbf{r}' - \mathbf{r}, \mathbf{R})], \\ -i\dot{h}_{\sigma_1,\sigma_2}(\mathbf{r}, \mathbf{R}) &= \hat{K}_{\sigma_1,\sigma_2}[h] \\ &\quad + \int d\mathbf{r}' [\tilde{\mathcal{E}}_{\sigma_1,s'}^*(\mathbf{r}', \mathbf{R}) p_{\sigma_2,s'}(-\mathbf{r} - \mathbf{r}', \mathbf{R}) \\ &\quad - \tilde{\mathcal{E}}_{\sigma_2,s'}(\mathbf{r}', \mathbf{R}) p_{\sigma_1,s'}^*(\mathbf{r} + \mathbf{r}', \mathbf{R})]. \end{aligned} \quad (37)$$

Here $\tilde{\mathcal{E}}_{\sigma,s}(\mathbf{r}, \mathbf{R}) = \mathcal{E}_{\sigma,s}(\mathbf{R})\delta(\mathbf{r}) - V(\mathbf{r})p_{\sigma,s}(\mathbf{r}, \mathbf{R})$ and the integrodifferential operators in Eq. (B7) are substituted by

$$\begin{aligned} \hat{H}_{s_1,s'} f_{s'} &= \left[-\frac{1}{2m_{s_1}} \frac{\partial^2}{\partial \mathbf{r}^2} + H_{s_1} \right] f_{s_1}(\mathbf{r}, \mathbf{R}) \\ &\quad - \int d\mathbf{r}' V(\mathbf{r}') e_{s_1,s'}(\mathbf{r}', \mathbf{R}) f_{s'}(\mathbf{r} - \mathbf{r}', \mathbf{R}), \\ \hat{H}_{\sigma_1,\sigma'} f_{\sigma'} &= -\frac{1}{2m_{\sigma_1,\sigma'}} \frac{\partial^2}{\partial \mathbf{r}^2} f_{\sigma'}(\mathbf{r}, \mathbf{R}) + H_{\sigma'} f_{\sigma_1}(\mathbf{r}, \mathbf{R}) \\ &\quad - \int d\mathbf{r}' V(\mathbf{r}') h_{\sigma_1,\sigma'}(\mathbf{r}', \mathbf{R}) f_{\sigma'}(\mathbf{r} - \mathbf{r}', \mathbf{R}). \end{aligned} \quad (38)$$

System (37) in the short-time limit (when all terms in the right-hand side but those depending on the external field are neglected) reproduces the results obtained in the Sec. II from Eq. (7). In particular the MWM response is obtained by the Fourier transform over the parameter \mathbf{R} similarly to Eq. (30). In what follows, we will use Eq. (37) in even simpler form assuming the single-pulse excitation only and thus omitting the dependence on \mathbf{R} . It suffices for our purpose of discussion of the effect of the internal dynamics and the Coulomb interaction. We also omit the off-diagonal elements of the

holes Hamiltonian $\hat{H}_{\sigma_1, \sigma_2}$ describing the mixing of the valence bands. Thus, we characterize these bands by m_{hh} and m_{lh} that is by the masses of the heavy and light holes, respectively.

Considering the terms besides the kinetic energy in the right-hand side of these equations as sources, the responses are governed by the free-particle propagators for the correlation functions and the Coulomb propagator for the interband polarization. The fundamental property of these propagators is that (in the short-time limit $t \rightarrow 0$) they turn to spatial δ functions owing to fast spatial oscillations of the kernel,

$$K(\mathbf{r}_1, \mathbf{r}_2; t) = \sqrt{\frac{m}{2\pi i t}} \exp[i m |\mathbf{r}_1 - \mathbf{r}_2|^2 / 2t]. \quad (39)$$

In order to evaluate the parameter m , we take into account that the evolution of the semiconductor—initially being in the ground state—is spanned by the spin classes $C_{1,2}$. Thus for the electron correlation function $m = \infty$, so that the propagator remains local. For the hole correlation functions, this parameter is finite for the off-diagonal elements $m = m_\Delta = (m_{lh}^{-1} - m_{hh}^{-1})^{-1}$. For the interband polarization in the short-time limit, the Coulomb propagator can be approximated by the free-particle propagator^{37,38} with $m \approx m_{xh} = (m_e^{-1} + m_{hh})^{-1}$ and $m \approx m_{xl} = (m_e^{-1} + m_{lh})^{-1}$, where $m_{x\sigma}$ are the excitons' reduced masses and m_e is the electron mass. The effect of the dispersion of the electrons and holes on the *exciton* polarization is expressed as a distortion of the exciton wave function under the action of the free-particle propagators. The time scale, at which the distortion becomes essential, can be estimated as the time required for the initial δ shape of the kernel to acquire the width of the order of the exciton Bohr radius. This leads to the estimate

$$t_c \lesssim \frac{m r_B^2}{4\pi}. \quad (40)$$

The numerical value of the typical time scale for GaAs is determined by $m_{hh} = 0.45m_0$, $m_{lh} = 0.082m_0$, $m = m_\Delta$, and $r_B = r_{xh} = \epsilon_b / 2m_{xh}e^2$, where m_0 is the electron mass in empty space and $\epsilon_b = 13$ is the background dielectric function. Using these values in Eq. (40) we find $t_c \sim 20$ fs. Thus, for pulses with duration shorter than 20 fs and the intensity sufficiently high to produce several Rabi flops, the approximation used in the Sec. II should be satisfactory. However, as will be shown below, the exciton polarizations corresponding to the states in the discrete spectrum are even less sensitive to the deviation of the dynamics from its short-time limit.

First we discuss the general effect of the electron-hole dispersion on the Rabi oscillations. For this we consider the case of excitation by a single circularly polarized pulse of high intensity. For such excitation, we neglect the contribution of the nonlinear Coulomb terms (see below) and additionally simplify the internal dynamics assuming the effects of the detuning and the energy offsets to be small at the time scales under consideration. Since in this case the translational invariance in the plane of the quantum well is restored, it is more convenient to solve Eq. (37)—rewriting it in the momentum representation.^{2,18,32} The Laplace transformation $p(\lambda) = \int_0^\infty p(t) e^{-\lambda t} dt$ of the solution is found to be

$$p_{\sigma,s}(\lambda, \mathbf{k}) = -i \frac{\mathcal{E}_{\sigma,s}}{(2\pi)^2} \cdot \frac{\lambda - i\omega_{\sigma,s}(\mathbf{k})}{\lambda[\lambda^2 + \omega_{\sigma,s}^2(\mathbf{k}) + \Omega_{\sigma,s}^2]}, \quad (41)$$

where $\omega_{\sigma,s}(\mathbf{k}) = k^2 / 2m_{x\sigma}$ and $\Omega_{\sigma,s}^2 = 4(|\mathcal{E}_{\sigma,s}|^2 + |\mathcal{E}_{\bar{\sigma},s}|^2)$ with $\bar{\sigma}$ being the hole spin state complimentary to σ in the respective spin class. In the case under consideration, because of circular polarization of the excitation pulse, only interband polarization corresponding to the same helicity is created, thus, if $\mathcal{E}_{\sigma,s} \neq 0$ then $\mathcal{E}_{\bar{\sigma},s} = 0$ and vice versa. We, however, write down the “full” expression for $\Omega_{\sigma,s}$ in order to show the relation with Eq. (12). The poles of the right-hand side of Eq. (41) determine the characteristic frequencies yielding the time dependence in the form

$$p_{\sigma,s}(t, \mathbf{k}) = -i \frac{\mathcal{E}_{\sigma,s}}{(2\pi)^2 \tilde{\Omega}_{\sigma,s}(\mathbf{k})} \left\{ \begin{aligned} & \sin[\tilde{\Omega}_{\sigma,s}(\mathbf{k})t] \\ & - 2i \frac{\omega_{\sigma,s}(\mathbf{k})}{\tilde{\Omega}_{\sigma,s}(\mathbf{k})} \sin^2[\tilde{\Omega}_{\sigma,s}(\mathbf{k})t/2] \end{aligned} \right\},$$

$$e_{s,s}(t, \mathbf{k}) = h_{\sigma,\sigma}(t, \mathbf{k}) = \frac{4|\mathcal{E}_{\sigma,s}^2}{(2\pi)^2 \tilde{\Omega}_{\sigma,s}^2(\mathbf{k})} \sin^2[\tilde{\Omega}_{\sigma,s}(\mathbf{k})t/2], \quad (42)$$

where $\tilde{\Omega}_{\sigma,s}(\mathbf{k}) = \sqrt{\Omega_{\sigma,s}^2 + \omega_{\sigma,s}^2(\mathbf{k})}$. It is interesting to note the similarity to the simple dynamical models discussed in Sec. I [see Eq. (42)].

The *exciton* polarization is found by convoluting Eq. (42) with the exciton wave function $\phi_{\sigma,s}(\mathbf{k}) = 2\sqrt{2\pi} r_{\sigma,s} (1 + k^2 r_{\sigma,s}^2)^{-3/2}$ according to Eq. (36). In Fig. 3 we plot the time dependence of the exciton polarization for different intensities of the external excitation. Since the exciton wave function is mostly localized inside the region $k \sim 1/r_B$ and by the assumption $\omega_{\sigma,s}(1/r_B) < \Omega_{\sigma,s}$, this convolution does not lead to appearance of additional frequencies, thus the oscillations of the exciton polarization are characterized by a slightly modified Rabi frequency $\Omega_{\sigma,s}$.

There are two significant corrections to the picture of the oscillations obtained in the Sec. II. First, there is slower than exponential decay due to dephasing of the components corresponding to different k . The decay can be estimated—considering the long time asymptotic $t \rightarrow \infty$. Since the phase in Eq. (42) reaches its stationary points at the ends of the k interval, i.e., at $k=0$, it implies the asymptotic power decay of the amplitude as $1/t$ independent on the intensity of the excitation. The second modification is related to the second term in the brackets in Eq. (42) [Fig. 3(b)]. This term has the form of oscillations with the Rabi frequency $\Omega_{\sigma,s}$ near a constant level, which position strongly depends on the intensity of the excitation for $\Omega_{\sigma,s} \lesssim 2\omega_{\sigma,s}(1/r_B)$ and slowly decays toward zero for higher values of the Rabi frequency as shown in Fig. 4.

Solutions (42) allow us to estimate the effect of the Coulomb interaction and to show that the contribution of the nonlinear terms into the right-hand side of equations of motion (37) vanishes with increasing the intensity of the excitation. Physically this is supported by (first) the observation that the interband polarization and the single-particle corre-

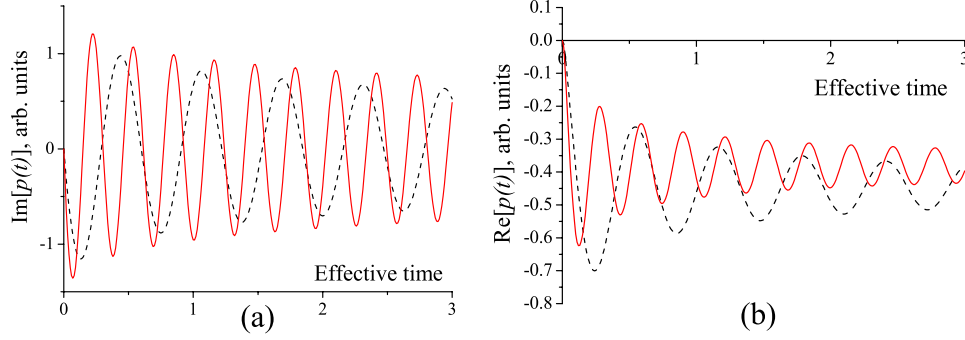


FIG. 3. (Color online) The effect of the electron-hole dispersion on the Rabi oscillations. The time dependence of the imaginary and the real parts of the exciton polarization is shown for $\Omega_{\sigma,s}/\omega_{\sigma,s}(1/r_B)=5$ (solid line) and $\Omega_{\sigma,s}/\omega_{\sigma,s}(1/r_B)=10$ (dashed line).

lation functions, as functions of the external excitation, are limited from above and, hence, the nonlinear terms are scaled as the exciton binding energy,²⁵ while the other terms increase with increasing the intensity of the external field. Second, one can also apply the result obtained from Eq. (B3) about vanishing the nonlinear terms in the limit of δ -functional profile of the interband polarization and the single-particle correlation functions. As follows from Eq. (42), with increasing the intensity of the excitation the spatial distribution of $p_{\sigma,s}$, $h_{\sigma,\sigma}$ and $e_{s,s}$ narrows with the characteristic width $\propto 1/k_\Omega$, where $k_\Omega = \sqrt{2m_{x\sigma}\Omega_{\sigma,s}}$.

There are different ways to estimate the contribution of the nonlinear Coulomb terms into the dynamics. In order to illustrate the application of the physical ideas mentioned above, we consider the contribution into the right-hand side of the equation of motion for the interband polarization coming from the term written in the second line in Eq. (B3). In the simplest case of a single-pulse excitation, this term yields an addition to the equation with respect to $p_{\sigma,s}(t, \mathbf{k})$ in the form

$$\Delta S_{\sigma,s}(\mathbf{k}) = \int d\mathbf{k}' V(\mathbf{k} - \mathbf{k}') [p_{\sigma,s}(\mathbf{k}') n(\mathbf{k}) - p_{\sigma,s}(\mathbf{k}) n(\mathbf{k}')], \quad (43)$$

where $n(\mathbf{x}) = e_{s,s}(\mathbf{x}) + h_{\sigma,\sigma}(\mathbf{x})$ and we have taken into account that the dynamics is restricted to a particular spin class. We

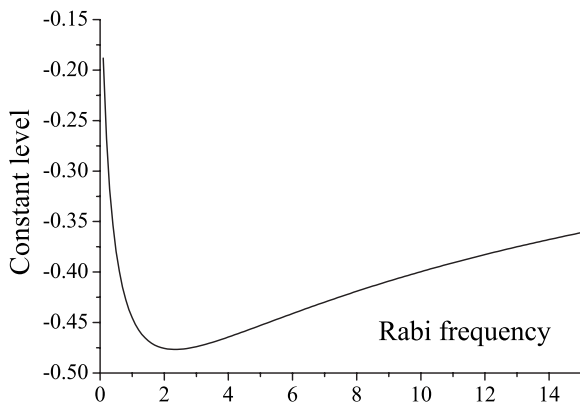


FIG. 4. The dependence of the constant level on the Rabi frequency $\Omega_{\sigma,s}/\omega_{\sigma,s}(1/r_B)$.

illustrate the typical line of calculations leaving only the first term in the expression for $p_{\sigma,s}(\mathbf{k})$ and approximating the oscillating terms by one, thus, presenting

$$\Delta S_{\sigma,s}(\mathbf{k}) \sim -i \frac{8\mathcal{E}_{\sigma,s}^3}{(2\pi)^4} \int d\mathbf{k}' \frac{V(\mathbf{k} - \mathbf{k}')}{\tilde{\Omega}_{\sigma,s}^2(\mathbf{k}) \tilde{\Omega}_{\sigma,s}^2(\mathbf{k}')} [\tilde{\Omega}_{\sigma,s}(\mathbf{k}) - \tilde{\Omega}_{\sigma,s}(\mathbf{k}')]. \quad (44)$$

Rescaling the wave vectors and using for the single-pulse excitation $\Omega_{\sigma,s} = 2\mathcal{E}_{\sigma,s}$, we obtain

$$\Delta S_{\sigma,s}(\mathbf{k}) \sim -i \frac{1}{(2\pi)^4} \sqrt{\frac{\epsilon_{x\sigma}}{\Omega_{\sigma,s}}} \zeta\left(\frac{\mathbf{k}}{k_\Omega}\right), \quad (45)$$

where $\epsilon_{x\sigma} = 2m_{x\sigma}e^4/\epsilon_b^2$ is the exciton binding energy and

$$\zeta(\boldsymbol{\kappa}) = \int d\boldsymbol{\kappa}' \frac{\sqrt{1 + \boldsymbol{\kappa}^4} - \sqrt{1 + \boldsymbol{\kappa}'^4}}{(1 + \boldsymbol{\kappa}^4)(1 + \boldsymbol{\kappa}'^4)|\boldsymbol{\kappa} - \boldsymbol{\kappa}'|}. \quad (46)$$

The dependence of this function on κ is shown in Fig. 5. It varies slowly over the interval $\kappa < 1/2$, where its magnitude is close to $\zeta(0) = \pi^2/\sqrt{2} - 8\Gamma^2(5/4)\sqrt{\pi} \approx -4.67$. Thus, if the Rabi frequency exceeds the exciton binding energy or (equivalently) if the typical spatial scale of the interband polarization $1/k_\Omega$ is smaller than the exciton Bohr radius, the effect of the nonlinear terms on the time dependence of the

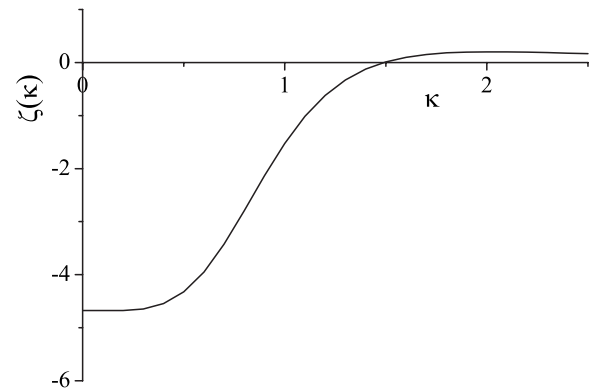


FIG. 5. The dependence on $\kappa = k/k_\Omega$ of the integral in Eq. (46) describing the effective modification of the source due to the nonlinear terms in the limit of high intensity of the external field.

exciton polarization reduces to a relatively small modification of the amplitude of the external field.

This argument allows one to reformulate the criterion of validity of the short-time approximation for the exciton polarization in terms of relation between the exciton Bohr radius r_B and the typical spatial width of the *interband* polarization due to the interaction with the external field $\sim 1/k_\Omega$. If $r_B k_\Omega \gg 1$, then the result of convolution of the exciton wave function with the interband polarization $p_{\sigma,s}$ does not depend on the details of the dependence of $p_{\sigma,s}(\mathbf{k})$ on \mathbf{k} . Indeed, since in this case the main contribution into the integral $P = \int d\mathbf{x} \psi^*(\mathbf{x}) p(\mathbf{x})$ comes from the small area near $\mathbf{x} = 0$, we can approximate the integral by

$$P \approx \psi^*(0) \int d\mathbf{x} p(\mathbf{x}) = (2\pi)^2 \psi^*(0) p(\mathbf{k} = 0), \quad (47)$$

which turns Eq. (42) into Eq. (17) exactly [including the cancelation of the factor $(2\pi)^2$]. This consideration shows that the exciton polarization corresponding to the states in the discrete spectrum is less sensitive to the deviations from the short-time dynamics of the system than the interband polarization. The latter contains information about all exciton states, including those from the continuum spectrum, whose spatial variation is no longer characterized by the exciton Bohr radius. The condition $r_B k_\Omega = \sqrt{(\Omega_{\sigma,s}/\epsilon_{x\sigma})} \gg 1$ imposes the limitation on the intensity of the external field that the period of the Rabi oscillations should be smaller than ~ 200 fs (for GaAs). It should be noted, however, that at this time scale more significant correction comes from the heavy-light hole beats with the characteristic period³⁹ ~ 100 fs, which impose stronger restriction on the intensity of the external excitation.

An interesting effect of the dispersion is revealed when the excitation pulse is not circularly polarized. In this case, both states constituting a spin class are involved in the dynamics resulting in the appearance of an additional characteristic frequency. In order to illustrate this effect, we consider the case when the intensity of the excitation is significantly high so that the effect dispersion of the hole-hole correlation function (which is quantified by the parameter $m_\Delta^{-1} = m_{lh}^{-1} - m_{hh}^{-1}$) can be treated perturbatively. In order to simplify the expression, we assume additionally that both heavy- and light-hole excitons are described by the same dispersion law $\omega_{\sigma,s}(\mathbf{k})$. Under these assumptions, the Laplace transform of the interband polarization can be written as

$$p_{\sigma,s}(\lambda, \mathbf{k}) = p_{\sigma,s}^{(0)}(\lambda, \mathbf{k}) + \frac{1}{2\pi^2 \lambda} \times \frac{\mathcal{E}_{\sigma,s} |\mathcal{E}_{\bar{\sigma},s}|^2 \omega_{\bar{\sigma},\sigma}(\mathbf{k})}{[\lambda^2 + \tilde{\Omega}_{\sigma,s}^2(\mathbf{k})][(\lambda + i\omega_{\sigma,s}(\mathbf{k})/2)^2 + \tilde{\Omega}_{\sigma,s}^2(\mathbf{k})/4]}, \quad (48)$$

where $p_{\sigma,s}^{(0)}(\lambda, \mathbf{k})$ is given by Eq. (41) and $\omega_{\bar{\sigma},\sigma}(\mathbf{k}) = k^2/2m_{\bar{\sigma},\sigma}$ with $m_{\bar{\sigma},\sigma}^{-1} = m_{\bar{\sigma}}^{-1} - m_{\sigma}^{-1}$, i.e., $|m_{\bar{\sigma},\sigma}| = m_\Delta$. The perturbation term in the right-hand side of Eq. (48) has poles at the same Rabi frequency $\tilde{\Omega}_{\sigma,s}^2(\mathbf{k})$, as in the case of the circularly polarized excitation and at the frequency $\approx \tilde{\Omega}_{\sigma,s}^2(\mathbf{k})/4$, yield-

ing a component of the exciton polarization oscillating with doubled period. We would like to emphasize that this frequency was initially present among the eigenfrequencies of the bright exciton classes [see Eq. (26)]. Its appearance, however, in the time evolution of the exciton polarization was prohibited in the dispersionless limit by the symmetry $p^{(2)*} \leftrightarrow p^{(1)}$ of dynamical equations [Eq. (22)]. The exciton finite masses break the symmetry allowing for the frequency $\Omega_{\sigma,s}^2(\mathbf{k})/4$ to contribute to the evolution. With this regard, it could be speculated that if the dark excitons would be involved in the dynamics their presence could be traced by the admixture of the frequencies specific for the dark exciton spin class C_3 [see Eq. (27)].

It should be emphasized that neither the effect of decay of the Rabi oscillations nor the admixture of “extra” frequencies are present in atomic analogs of the semiconductor Rabi oscillations. The decay is essentially caused by the spatial spread of the true (single-particle) eigenstates of the system. The appearance of additional frequencies is related to the existence of different kinds of holes with continuous spectra characterized by different masses.

IV. CONCLUSION

We have studied the short-time semiconductor response with respect to resonant high intensity excitations. The main results are obtained neglecting the slow dynamics that is on the time scale provided by detuning. We have calculated exactly the excitation polarization $P_\mu = \langle \psi(t) | B_\mu | \psi(t) \rangle$ in the coherent limit. We have found that if the semiconductor is excited by a field consisting of one or two plane waves, the polarization of bright excitons demonstrates Rabi oscillations, while the polarization of dark excitons is identically zero. The main difference between the single-wave and two-wave excitations is that in the latter case, the Rabi oscillations of the multiwave mixing (MWM) polarizations are generally a superposition of two harmonics ($\Omega_1 > \Omega_2$), unless the amplitudes of the waves satisfy a special relation. For example, if both waves are circularly polarized with the same helicity then $\Omega_1 - \Omega_2 \sim 2dE$, where E is amplitude of the weaker wave and the slow harmonic Ω_2 collapses only if the amplitudes of both waves are the same. Also, in the case of the two-pulse excitation, oscillations of the exciton polarization decay $\propto t^{-1/2}$ owing to redistribution of the excitation over the MWM polarizations.

In order to establish the dynamical origin of the vanishing polarization of the dark states, we considered the operator dynamics governed by the external field. We found that there exist three invariant spin classes, which do not mix with the evolution of the system. Two of these classes correspond to bright excitons and the third one contains all the dark states. It has turned out that the operator dynamics is described by six frequencies. Only two of those frequencies show up as the Rabi frequencies in the time evolution of the exciton polarizations (one for each bright classes), while others are prohibited by a hidden symmetry.

The short-time approximation used in the first part of the paper appears naturally in the more general context of the semiconductor Bloch equation (SBE). We use the SBE in

order to discuss the effect of the Coulomb interaction and the effect of the continuous spectra of the electrons and holes on the results obtained in the limit of immediate response with respect to intensive excitation. We found that if the system is initially in the ground state, then the SBE preserves the invariant spin classes. Thus, it proves that as long as the SBE holds and the approximation of angular independent dipole moment is justified the dark excitons do not appear in the dynamics.

An interesting result of studying the short-time approximation within the context of the SBE is that the Coulomb interaction does not lead to essential changes if the intensity of excitation is sufficiently high compared to the exciton binding energy. Moreover, including the nonlinear exchange terms directly to the equations of the short-time limit (i.e., without the kinetic energy) adds nothing since they cancel each other. In turn, the effects of the continuous spectrum of the electrons and holes are much more significant. We show that the electron-hole dispersion, while does not change significantly, the frequency of the oscillations of the exciton polarization leads to the power-law decay of the oscillations. An interesting effect is related to the mass difference between heavy and light holes. It breaks the symmetry that selects only two Rabi frequencies out of six that are introduced into the system by the external field and leads to the admixture of the second nontrivial frequency for a specific bright class.

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APPENDIX A: HEISENBERG REPRESENTATION INDUCED BY THE EXTERNAL FIELD

In order to derive Eq. (21) one can either consider a regularization of the δ function, which appears in the anticommutation relations and in the right-hand side of Eq. (19) or consider a more general form of Eq. (19) with the amplitudes being scalar functions, which are convoluted with the electron and hole operators,

$$\hat{F}(t) = A(t) + \sum_{\sigma_1, \sigma_2} \int dx dy g_{\sigma_1, \sigma_2}(\mathbf{x}, \mathbf{y}; t) v_{\sigma_1}^\dagger(\mathbf{x}) v_{\sigma_2}(\mathbf{y}) + \dots \quad (\text{A1})$$

Furthermore, the Heisenberg equation of motion produces the dynamical equations with respect to the amplitudes,

$$\dot{f}_{s_1, s_2}(\mathbf{x}, \mathbf{y}) = i\mathcal{E}_{\sigma', s_1}(\mathbf{x}) \pi_{\sigma', s_2}^{(2)}(\mathbf{x}, \mathbf{y}) - i\pi_{\sigma', s_1}^{(1)}(\mathbf{y}, \mathbf{x}) \mathcal{E}_{\sigma', s_2}^*(\mathbf{y}),$$

$$\dot{g}_{\sigma_1, \sigma_2}(\mathbf{x}, \mathbf{y}) = i\mathcal{E}_{\sigma_1, s'}(\mathbf{x}) \pi_{\sigma_2, s'}^{(2)}(\mathbf{y}, \mathbf{x}) - i\pi_{\sigma_1, s'}^{(1)}(\mathbf{x}, \mathbf{y}) \mathcal{E}_{\sigma_2, s'}^*(\mathbf{y}),$$

$$\dot{\pi}_{\sigma, s}^{(1)}(\mathbf{x}, \mathbf{y}) = -i\mathcal{E}_{\sigma, s'}(\mathbf{x}) f_{s, s'}(\mathbf{y}, \mathbf{x}) - i g_{\sigma, \sigma'}(\mathbf{x}, \mathbf{y}) \mathcal{E}_{\sigma', s}(\mathbf{y}),$$

$$\dot{\pi}_{\sigma, s}^{(2)}(\mathbf{x}, \mathbf{y}) = i\mathcal{E}_{\sigma, s'}^*(\mathbf{x}) f_{s', s}(\mathbf{x}, \mathbf{y}) + i g_{\sigma', \sigma}(\mathbf{y}, \mathbf{x}) \mathcal{E}_{\sigma', s}^*(\mathbf{y}),$$

$$\dot{A} = i \int d\mathbf{x} [\pi_{\sigma', s'}^{(1)}(\mathbf{x}, \mathbf{x}) \mathcal{E}_{\sigma', s'}^*(\mathbf{x}) - \pi_{\sigma', s'}^{(2)}(\mathbf{x}, \mathbf{x}) \mathcal{E}_{\sigma', s'}(\mathbf{x})], \quad (\text{A2})$$

where the summation over dashed spin variables is implied. The last equation gives the time dependence of $\langle \hat{F}(t) \rangle$ [compare with Eq. (11)]. Considering the dynamics of the diagonal $\mathbf{x}=\mathbf{y}$ values of the amplitudes in the case of spatially homogeneous excitation $\mathcal{E}_{\sigma, s}(\mathbf{x}) = \mathcal{E}_{\sigma, s}$, one obtains Eq. (21).

It should be emphasized that Eq. (A2) is not the semiconductor Bloch equation (SBE). Equation (A2) is written with respect to amplitudes of different operators entering the Heisenberg representation of the operator $\hat{F}(t)$. The SBE, in turn, is written for average values of the specific operators [see, e.g., Eq. (B5)]. While Eq. (A2) and the SBE have a similar structure (in the linear approximation for the SBE), they are different as can be seen comparing closely the spin indices in Eqs. (A2) and (B5). In order to find an average value of a particular operator using Eq. (A2), one needs first to solve these equations for specific initial conditions and then use this solution in the last equation of Eq. (A2).

APPENDIX B: SEMICONDUCTOR BLOCH EQUATION IN THE COORDINATE REPRESENTATION

In the main text we consider the semiconductor response under the action of an external field, which is spatially inhomogeneous in the plane of the quantum well. Because the system does not possess the translational invariance, it is convenient to use the SBE in the coordinate representation.

The Hamiltonian of an excited semiconductor in the rotating wave approximation has the form

$$H = H_{el} + H_h + H_C + H_e, \quad (\text{B1})$$

where H_{el} and H_h are the standard electron and hole single-particle Hamiltonians,^{2,18} H_C is the Hamiltonian of the Coulomb interaction, and H_e describes the light-matter interaction

$$H_e = \int d\mathbf{x} \mathcal{E}_{\sigma, s}(\mathbf{x}) v_{\sigma}^\dagger(\mathbf{x}) c_s^\dagger(\mathbf{x}) + \text{H.c.}, \quad (\text{B2})$$

where $\mathcal{E}_{\sigma, s}(\mathbf{x}) = \langle s | \mathbf{p} | \sigma \rangle \cdot \mathbf{E}(\mathbf{x})$, operator $v_{\sigma}^\dagger(\mathbf{x})$ creates a hole with the spin state σ at point \mathbf{x} , and $c_s^\dagger(\mathbf{x})$ is the respective electron creating operator. The equations of motion for the interband polarizations $p_{\sigma, s}(\mathbf{x}_1, \mathbf{x}_2) = \langle c_s(\mathbf{x}_2) v_{\sigma}(\mathbf{x}_1) \rangle$, the electron-electron $e_{s_1, s_2}(\mathbf{x}_1, \mathbf{x}_2) = \langle c_{s_1}^\dagger(\mathbf{x}_1) c_{s_2}(\mathbf{x}_2) \rangle$, and the hole-hole $h_{\sigma_1, \sigma_2}(\mathbf{x}_1, \mathbf{x}_2) = \langle v_{\sigma_1}^\dagger(\mathbf{x}_1) v_{\sigma_2}(\mathbf{x}_2) \rangle$ correlation functions are derived from the equation of motion $d\langle f \rangle / dt = i\langle [H, f] \rangle$ using the Hartree-Fock approximation for the terms describing the Coulomb interaction.

Straightforward calculations give immediately quite cumbersome system of equations. We provide explicitly only one equation, which shows the structure of the nonlinear Coulomb terms,

$$\begin{aligned}
i\dot{p}_{\sigma,s}(\mathbf{x}_1, \mathbf{x}_2) &= K_{\sigma,s}[p] + [U_{\Delta}(\mathbf{x}_1) - U_{\Delta}(\mathbf{x}_2)]p_{\sigma,s}(\mathbf{x}_1, \mathbf{x}_2) \\
&\quad - \int d\mathbf{x}' [V(\mathbf{x}_1 - \mathbf{x}') - V(\mathbf{x}_2 - \mathbf{x}')] \\
&\quad \times [h_{\sigma',\sigma}(\mathbf{x}', \mathbf{x}_1)p_{\sigma',s}(\mathbf{x}', \mathbf{x}_2) \\
&\quad - p_{\sigma,s'}(\mathbf{x}_1, \mathbf{x}')e_{s',s}(\mathbf{x}', \mathbf{x}_2)] + \mathcal{E}_{\sigma,s}(\mathbf{x}_1)\delta(\mathbf{x}_1 - \mathbf{x}_2) \\
&\quad - h_{\sigma',\sigma}(\mathbf{x}_2, \mathbf{x}_1)\mathcal{E}_{\sigma',s}(\mathbf{x}_2) - \mathcal{E}_{\sigma,s'}(\mathbf{x}_1)e_{s',s}(\mathbf{x}_1, \mathbf{x}_2).
\end{aligned} \tag{B3}$$

Here and below the summation over dashed spin indices is implied. This equation clearly demonstrates that at the diagonal $\mathbf{x}_1 = \mathbf{x}_2$, the contributions of the Coulomb terms cancel each other. In Eq. (B3) $U_{\Delta}(\mathbf{x})$ describes an effective background potential created by the local imbalance between the electrons and holes,

$$U_{\Delta}(\mathbf{x}) = \int d\mathbf{x}' V(\mathbf{x} - \mathbf{x}') [e_{s',s'}(\mathbf{x}', \mathbf{x}') - h_{\sigma',\sigma'}(\mathbf{x}', \mathbf{x}')]. \tag{B4}$$

For a semiconductor excited by a single plane wave, this potential vanishes in an overall neutral system due to the translational invariance. In the two wave excitation setup, the effect of U_{Δ} is small for bound exciton states if the order of the MWM response is not too high.

The dynamical equations significantly simplify after the terms being rearranged to form renormalizations of the single-particle energies and the coupling between the charge densities and the interband polarizations,

$$\begin{aligned}
i\dot{p}_{\sigma,s}(\mathbf{x}_1, \mathbf{x}_2) &= \hat{K}_{\sigma,s}[p] + \mathcal{E}_{\sigma,s}(\mathbf{x}_1)\delta(\mathbf{x}_1 - \mathbf{x}_2) \\
&\quad - \int d\mathbf{x}' [\tilde{\mathcal{E}}_{\sigma',s}(\mathbf{x}', \mathbf{x}_2)h_{\sigma',\sigma}(\mathbf{x}', \mathbf{x}_1) \\
&\quad - \tilde{\mathcal{E}}_{\sigma,s'}(\mathbf{x}', \mathbf{x}_1)e_{s',s}(\mathbf{x}', \mathbf{x}_2)],
\end{aligned}$$

$$\begin{aligned}
-i\dot{e}_{s_1,s_2}(\mathbf{x}_1, \mathbf{x}_2) &= \hat{K}_{s_1,s_2}[e] + \int d\mathbf{x}' [\tilde{\mathcal{E}}_{\sigma',s_1}^*(\mathbf{x}', \mathbf{x}_1)p_{\sigma',s_2}(\mathbf{x}', \mathbf{x}_2) \\
&\quad - \tilde{\mathcal{E}}_{\sigma',s_2}(\mathbf{x}', \mathbf{x}_2)p_{\sigma',s_1}^*(\mathbf{x}', \mathbf{x}_1)],
\end{aligned}$$

$$\begin{aligned}
-i\dot{h}_{\sigma_1,\sigma_2}(\mathbf{x}_1, \mathbf{x}_2) &= \hat{K}_{\sigma_1,\sigma_2}[h] + \int d\mathbf{x}' [\tilde{\mathcal{E}}_{\sigma_1,s'}^*(\mathbf{x}_1, \mathbf{x}')p_{\sigma_2,s'}(\mathbf{x}_2, \mathbf{x}') \\
&\quad - \tilde{\mathcal{E}}_{\sigma_2,s'}(\mathbf{x}_2, \mathbf{x}')p_{\sigma_1,s'}^*(\mathbf{x}_1, \mathbf{x}')],
\end{aligned} \tag{B5}$$

where we have introduced the modified coupling between the interband polarization and the charge densities,

$$\tilde{\mathcal{E}}_{\sigma,s}(\mathbf{x}, \mathbf{y}) = \mathcal{E}_{\sigma,s}(\mathbf{x})\delta(\mathbf{x} - \mathbf{y}) - V(\mathbf{x} - \mathbf{y})p_{\sigma,s}(\mathbf{x}, \mathbf{y}). \tag{B6}$$

We represent the time dependence of the external field in the form $E(t) = \tilde{E}(t)e^{-i\Omega t}$, where the amplitude $\tilde{E}(t)$ is assumed to be constant during the excitation. In the rotating frame, the operators

$$\begin{aligned}
\hat{K}_{\sigma,s}[p] &= \hat{H}_{\sigma,\sigma'}p_{\sigma',s} + p_{\sigma,s}\hat{H}_{s',s} - V(\mathbf{x}_1 - \mathbf{x}_2)p_{\sigma,s}(\mathbf{x}_1, \mathbf{x}_2) \\
&\quad - \Omega p_{\sigma,s}(\mathbf{x}_1, \mathbf{x}_2), \\
\hat{K}_{s_1,s_2}[e] &= \hat{H}_{s_1,s'}e_{s',s_2} - e_{s_1,s'}\hat{H}_{s',s_2}, \\
\hat{K}_{\sigma_1,\sigma_2}[h] &= \hat{H}_{\sigma_1,\sigma'}h_{\sigma',\sigma_2} - h_{\sigma_1,\sigma'}\hat{H}_{\sigma',\sigma_2}
\end{aligned} \tag{B7}$$

are expressed in terms of the integrodifferential operators \hat{H} . The action of these operators is defined by

$$\begin{aligned}
\hat{H}_{s_1,s'}f_{s'} &= \left[-\frac{1}{2m_{s_1}}\frac{\partial^2}{\partial \mathbf{x}_1^2} + V_e(\mathbf{x}_1) + U_{\Delta}(\mathbf{x}_1) + H_{s_1} \right] f_{s_1}(\mathbf{x}_1, \mathbf{x}_2) \\
&\quad - \int d\mathbf{x}' V(\mathbf{x}_1 - \mathbf{x}')e_{s_1,s'}(\mathbf{x}_1, \mathbf{x}')f_{s'}(\mathbf{x}', \mathbf{x}_2), \\
\hat{H}_{\sigma_1,\sigma'}f_{\sigma'} &= -\frac{1}{2m_{\sigma_1,\sigma'}}\frac{\partial^2}{\partial \mathbf{x}_1^2}f_{\sigma'}(\mathbf{x}_1, \mathbf{x}_2) + [V_h(\mathbf{x}_1) \\
&\quad - U_{\Delta}(\mathbf{x}_1) + H_{\sigma_1}]f_{\sigma_1}(\mathbf{x}_1, \mathbf{x}_2) \\
&\quad - \int d\mathbf{x}' V(\mathbf{x}_1 - \mathbf{x}')h_{\sigma_1,\sigma'}(\mathbf{x}_1, \mathbf{x}')f_{\sigma'}(\mathbf{x}', \mathbf{x}_2),
\end{aligned} \tag{B8}$$

with V_e and V_h being the electron and hole confinement potentials, which effect on the spatial motion of the electrons and holes is estimated by the δ -functional approximation for the quantum well. The parameters H_s and H_{σ} denote the energy offsets of the respective bands. The diagonal elements of m_{σ_1,σ_2} are the masses of the light and heavy holes, while the off-diagonal elements account for the valence-bands mixing.

Thus, $\hat{K}_{\sigma,s}$ has the meaning of a two-particle Hamiltonian with the Coulomb interaction between them, while the properties of free particles are described by the Hamiltonians $\hat{H}_{\sigma_1,\sigma_2}$ and \hat{H}_{s_1,s_2} .

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