

Nonperturbative phenomena in semiconductor four-wave mixing spectra

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Nonperturbative phenomena in four-wave mixing spectra of semiconductors are studied using the exact solution of a widely used phenomenological nonlinear equation of motion of the exciton polarization. It is shown that the Coulomb interaction, included in the nonlinearity, leads to two characteristic effects, which are essentially of dynamical origin—a split of the exciton peak and a nonmonotonous dependence of the response at the exciton frequency on the magnitude of the external field. Relations between the spectral features and the parameters of the system are obtained. It is found that the transition from perturbative to nonperturbative regimes is controlled by parameters inversely proportional to the decay rate. It implies that the condition of low excitation density does not necessarily warrant applicability of the perturbational approach.

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I. INTRODUCTION

One of the clearest manifestations of the many-body effects in semiconductors is the phenomenon of the nonlinear optical response. The Pauli blocking and the Coulomb interaction between the quasiparticles lead to the dependence of the time evolution of the exciton polarization on its spatial distribution.^{1–8} When a semiconductor quantum well is excited by two successive pulses, they produce the signal in directions which are prohibited in the linear regime while still conserve momentum in four-wave and multiwave mixing. The dynamical origin⁹ of the formation of the four-wave mixing (FWM) signal is naturally incorporated into the description in terms of the exciton modes characterized by the frequency ω_0 and the in-plane wave vector \mathbf{k} . From this perspective the effects of the Pauli blocking and of the Coulomb interaction are clearly different. The Pauli blocking reduces *locally* the intensity of the field-matter interaction according to the magnitude of the local polarization. As a result, the excitation field with particular value of the wave-vector \mathbf{q} becomes coupled to the exciton modes with different $\mathbf{k} \neq \mathbf{q}$. The Coulomb interaction between the excitons leads to the direct coupling between the exciton modes. In particular, when two modes with \mathbf{k}_1 and \mathbf{k}_2 are excited, the relevant coupled modes are characterized by $\mathbf{k}^{(n)} = \mathbf{k}_2 + n\Delta\mathbf{k}$, where $-\infty < n < \infty$ is an integer number and $\Delta\mathbf{k} = \mathbf{k}_1 - \mathbf{k}_2$. Although initially all energy is concentrated in the modes $\mathbf{k}^{(0)}$ and $\mathbf{k}^{(1)}$ during the evolution the energy is redistributed among the coupled modes. In particular it leads to the formation of the FWM signals, which correspond to $n=2$ and $n=-1$.

It follows from this picture that the redistribution of the energy between the modes characterized by different \mathbf{k} should not be the only manifestation of the mode coupling. This coupling should also lead to the modification of the frequencies of the exciton modes. Indeed, without the interaction and neglecting the dispersion of the exciton modes one has a many-fold degeneracy at the exciton resonance frequency ω_0 . The interaction between the modes should lift the degeneracy and, if the excitation is sufficiently strong, make the split of the exciton peak visible in the FWM spectrum.

II. NONLINEAR DYNAMICS OF THE EXCITON POLARIZATION

The effect of the modification of the frequencies of the exciton modes caused by the coupling is a nonperturbative effect.^{10,11} In order to study the problem we present the dynamics of the exciton polarization as the exact solution to the phenomenological nonlinear equation,¹²

$$\begin{aligned} \dot{P}(t; \mathbf{r}) = & -(i\omega_0 + \gamma)P(t; \mathbf{r}) - i\beta|P(t; \mathbf{r})|^2P(t; \mathbf{r}) \\ & - i\Omega_R(t; \mathbf{r})[1 - |P(t; \mathbf{r})|^2/P_{\text{sat}}^2], \end{aligned} \quad (1)$$

where ω_0 is the detuning, i.e., the difference between the frequency of the external field and the exciton frequency in the stationary frame, γ is the phenomenological decay rate, P_{sat}^2 is the exciton saturation density, and $\beta = \beta' - i\beta''$, with β' , $\beta'' \geq 0$, is a phenomenological parameter quantifying the interaction between the excitons. The positive real and negative imaginary parts of this parameter constitute the excitation induced shift¹⁰ (EIS) and the excitation induced decay^{13–15} (EID), respectively. The interaction with the external field is quantified by the Rabi frequency $\Omega_R(t; \mathbf{r}) = dE(t; \mathbf{r})$, where $E(t; \mathbf{r})$ is the envelope of the external field and d is the respective dipole moment. Writing Eq. (1) we have taken into account that the external field is coupled only to the exciton s states and have normalized the exciton polarization by the value of the exciton wave function at the origin, $\phi^*(0) = 2\sqrt{2}/\pi a_B^2$, with a_B being the exciton Bohr radius.

Phenomenologically Eq. (1) has provided valuable interpretations for nonlinear measurements.^{12,16,17} The nonlinear term $\propto \beta$ may be viewed as the short-time limit of the memory function,³ which is exact to the third order in the exciting electric field.¹⁸ The nonperturbative solutions have been considered in Refs. 19 and 20 and many papers referenced in Ref. 11. Equation (1) takes into account only the exciton states and assumes that the effect of biexcitons is rather small. The possibility to neglect the biexciton contribution is supported by the observation that the true bound

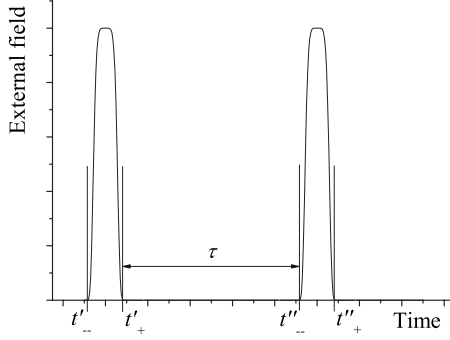


FIG. 1. The semiconductor is excited by two short pulses separated by the delay time $\tau = t''_- - t'_+$.

two-exciton states, whose effect would be most significant, do not form under the excitation with definite helicity. Also, as we will see later the main frequency region of interest is limited from below by the frequency of the exciton resonance, which makes it spectrally resolved from the frequencies where the biexciton effect would be the most significant.

We consider the excitation of the semiconductor by two short circularly polarized pulses with the same helicity (see Fig. 1) acting on the system during the intervals $t'_- < t < t'_+$ and $t''_- < t < t''_+$. Between the pulses, $t'_+ < t < t''_-$ and after the second pulse $t > t''_+$ the dynamics of the exciton polarization is free and satisfies Eq. (1) with $E=0$. We study the free dynamics polarization $P_0(t)$ before calculating the polarization $P_{1,2}(t)$ of the driven dynamics. Then we are going to solve for the polarization in the following order: $P_1(t)$ between $t'_- < t < t'_+$, $P_{10}(t)$ between $t'_+ < t < t''_-$, $P_2(t)$ between $t''_- < t < t''_+$, and at the end the final solution $P_{20}(t)$ for $t > t''_+$.

The free dynamics [$\Omega_R(t)=0$] can be solved exactly by noting that Eq. (1) in this regime is reduced to the simple form

$$\dot{P}_0(t; \mathbf{r}) = -[i\omega_0 + \gamma + i\Omega(t; \mathbf{r})]P_0(t; \mathbf{r}), \quad (2)$$

where $\Omega(t; \mathbf{r}) = \beta|P_0(t; \mathbf{r})|^2$, and by observing that Eq. (1) yields an equation for the magnitude of the polarization,

$$\frac{d}{dt}|P_0(t; \mathbf{r})|^2 = -2[\gamma + \beta''|P_0(t; \mathbf{r})|^2]|P_0(t; \mathbf{r})|^2 \quad (3)$$

with the solution

$$|P_0(t; \mathbf{r})| = |P_0(0; \mathbf{r})|e^{-\gamma t}A[t; |P_0(0; \mathbf{r})|], \quad (4)$$

where

$$A^2[t; |P_0(0; \mathbf{r})|] = \left[1 + \frac{\beta''|P_0(0; \mathbf{r})|^2}{\gamma}(1 - e^{-2\gamma t}) \right]^{-1}. \quad (5)$$

The free propagator may be expressed in terms of the amplitude modulation $A(t; \mathbf{r})$ and the phase modulation $\varphi(t; \mathbf{r})$,

$$\begin{aligned} P_0(t; \mathbf{r})/P_0(0; \mathbf{r}) &= \theta[t; |P_0(0; \mathbf{r})|] \\ &= A[t; |P_0(0; \mathbf{r})|]e^{-i\omega_0 t - \gamma t + i\varphi(t; \mathbf{r})}, \end{aligned} \quad (6)$$

where

$$\varphi(t; \mathbf{r}) = \frac{\beta'}{\beta''} \ln A[t; |P_0(0; \mathbf{r})|]. \quad (7)$$

The amplitude term shows the nonperturbative effect of EID. It is interesting to note that EID does not lead to a mere modification of the decay rate, γ . Instead the amplitude modulation $A(t; \mathbf{r})$ decreases to a fraction of the initial amplitude at twice the linear rate, i.e., 2γ . The phase modulation shows an oscillation dependent on the nonlinear quality factor β'/β'' . This is related to the Goldstone mode in the excitons studied in Ref. 21.

The initial conditions for Eq. (2) are determined by the polarization distribution right after the external field is switched off. We find the immediate response of the system assuming that the duration of the excitation pulses is much shorter than the typical dynamical time scales determined by detuning and the decay rate.

We turn now to the driven [$\Omega_R(t) \neq 0$] time evolution of the polarization; i.e., we consider the time interval $t_- < t < t_+$ where the particular excitation pulse does not vanish and factor out the term $\exp(i\mathbf{k} \cdot \mathbf{r})$ so that we can consider the excitation pulse to be spatially homogeneous. Neglecting the contribution to the phase $\sim \int_{t_-}^{t_+} [i\omega_0 + \gamma + i\Omega(t; \mathbf{r})]P_{1,2}(t; \mathbf{r})dt$ we solve the dynamical equation and find the relation between the polarization at the instances t_- and t_+ ,

$$\begin{aligned} P_{1,2}(t_+) &= P'_{1,2}(t_-) \\ &- i\sqrt{P_{\text{sat}}^2 - P_{1,2}^{\prime 2}(t_-)} \tanh \left\{ \sqrt{P_{\text{sat}}^2 - P_{1,2}^{\prime 2}(t_-)} \frac{\epsilon}{P_{\text{sat}}^2} \right. \\ &\left. - \operatorname{arctanh} \left[\frac{P'_{1,2}(t_-)}{\sqrt{P_{\text{sat}}^2 - P_{1,2}^{\prime 2}(t_-)}} \right] \right\}, \end{aligned} \quad (8)$$

where $P'_{1,2}(t_-) = \operatorname{Re}[P_{1,2}(t_-)]$ and $P''_{1,2}(t_-) = \operatorname{Im}[P_{1,2}(t_-)]$, and we have introduced the area of the exciting pulse $\epsilon = \int dt \Omega_R(t)$.

Using the solutions for the free and driven polarization in Eqs. (2) and (8), we solve the time evolution of the polarization in all the four regions $t'_- < t < t'_+$, $t'_+ < t < t''_-$, $t''_- < t < t''_+$, and $t > t''_+$. Assuming that the system initially is in the ground state $P(t_-) \equiv 0$ we find the exciton polarization created by the first pulse,

$$P_1(t'_+; \mathbf{r}) = -ie^{i\mathbf{k}_1 \cdot \mathbf{r}} P_1(t'_+), \quad (9)$$

where

$$P_1(t'_+) = P_{\text{sat}} \tanh \left(\frac{\epsilon}{P_{\text{sat}}} \right). \quad (10)$$

It follows from this equation that the saturation effect for the first pulse reduces to a simple (although nonlinear) renormalization of the pulse area. Using Eq. (9) in Eq. (4) we can see that the effective frequency $\Omega(t; \mathbf{r})$ determining the dynamics of the polarization is constant across the sample. As a result there is no coupling between the exciton modes characterized by different \mathbf{k} .

Denoting the delay time, the time separation between the pulses, by $\tau = t''_- - t'_+$, we obtain the polarization right before the arrival of the second pulse,

$$P_{10}(t''_+; \mathbf{r}) = P_1(t'_+; \mathbf{r}) \theta_{10}(\tau), \quad (11)$$

where $\theta_{10}(\tau) = \theta[\tau; |P_1(t'_+)|]$. The spatial distribution of the polarization $P_{10}(t''_+; \mathbf{r})$ plays the role of the initial condition for the immediate response with respect to the second pulse according to Eq. (8). For the analysis of the time evolution of the polarization during the second pulse it is convenient to factor $\exp(i\mathbf{k}_2 \cdot \mathbf{r})$ out of $P_2(t; \mathbf{r})$ introducing

$$\tilde{P}_2(t; \Delta \mathbf{k} \cdot \mathbf{r}) = e^{-i\mathbf{k}_2 \cdot \mathbf{r}} P_2(t; \mathbf{r}) \quad (12)$$

with $\Delta \mathbf{k} = \mathbf{k}_1 - \mathbf{k}_2$. The reduced distribution $\tilde{P}_2(t; \Delta \mathbf{k} \cdot \mathbf{r})$ satisfies Eq. (1) with the modified spatial profile of the external field $\tilde{\Omega}_R(t) = e^{-i\mathbf{k}_2 \cdot \mathbf{r}} \Omega_R(t; \mathbf{r})$. The form of the initial conditions in Eq. (8) changes according to

$$\tilde{P}_{10}(t''_+; \Delta \mathbf{k} \cdot \mathbf{r}) = -i \theta_{10}(\tau) P_1(t'_+) e^{i\Delta \mathbf{k} \cdot \mathbf{r}}. \quad (13)$$

It follows from Eqs. (8) and (13) that for the second pulse the role of the saturation effect is twofold. It modifies the pulse area and excites all modes $\mathbf{k}^{(n)}$ rather than just a single mode as we had for the first pulse. We present the polarization as a superposition of the multiwave mixing modes,

$$\tilde{P}_2(t; \kappa) = \sum_n P_2^{(n)}(t) e^{i n \kappa}, \quad (14)$$

where $\kappa = \Delta \mathbf{k} \cdot \mathbf{r}$ and $P_2^{(n)}(t)$ are the amplitudes of the multiwave mixing polarizations. Substituting this representation into Eq. (2) we can see that in terms of the multiwave mixing modes the free dynamics of the polarization can be presented as the dynamics of a system with an infinite number of degrees of freedom coupled to each other due to the nonlinearity,

$$\frac{d}{dt} P_2^{(n)}(t) = -(i\omega_0 + \gamma) P_2^{(n)}(t) - i \sum_m \Omega_{n-m}(t) P_2^{(m)}(t), \quad (15)$$

where $\Omega_n(t) = \beta(2\pi)^{-1} \int_{-\pi}^{\pi} |\tilde{P}_2(t; \kappa)|^2 e^{-i\kappa n} d\kappa$. The initial conditions for Eq. (15) are constituted by the immediate response to the second pulse, thanks to the saturation effect. This picture clearly illustrates the difference between the effect of the Pauli blocking and the Coulomb interaction on formation of the multiwave mixing response.

Using the solution of the equation of motion for free polarization dynamics we find

$$P_2^{(n)}(t) = \frac{1}{2\pi} \int_{-\pi}^{\pi} \tilde{P}_2(t''_+; \kappa) \theta[\tau; |\tilde{P}_2(t''_+; \kappa)|] e^{-i\kappa n} d\kappa. \quad (16)$$

This equation and Eqs. (8) and (13) give the exact evolution of the exciton polarization in the limit of short excitation pulses in the two-pulse scheme. Formally, one can obtain from Eq. (16) the spectrum $P_2^{(n)}(\omega)$ using the Fourier transform of $\theta[\tau; |\tilde{P}_2(t''_+; \kappa)|]$ with respect to time,

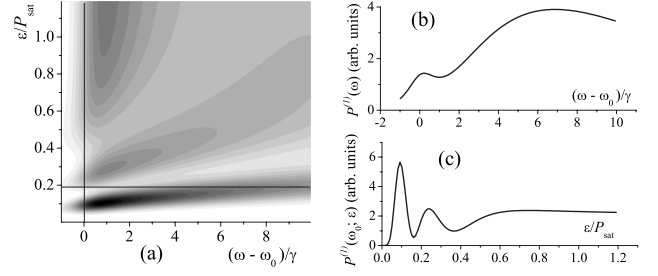


FIG. 2. (a) Four-wave mixing spectrum corresponding to $2\mathbf{k}_1 - \mathbf{k}_2$ as a function of the normalized pulse area. The parameters of the system are chosen to be $\beta'/\gamma = 5 \times 10^2$, $\beta'/2\beta'' = 5$. The horizontal and the vertical lines show the positions of the sections of the surface $P^{(1)}(\omega; \epsilon)$ presented in (b) and (c). (b) The FWM spectrum at the fixed value $\epsilon/P_{\text{sat}} = 0.19$. (c) The dependence of the FWM response at the exciton frequency $P^{(1)}(\omega_0; \epsilon)$ as a function of the pulse area.

$$\theta[\omega; |\tilde{P}_2(t''_+; \kappa)|] = \frac{i}{\gamma(w + i/2)} {}_2F_1 \left[1; \frac{1}{2} + iX; \frac{3}{2} - iw; -\frac{\beta'' |\tilde{P}_2(t''_+; \kappa)|^2}{\gamma} \right], \quad (17)$$

where $w = (\omega - \omega_0)/2\gamma$, $X = \beta'/2\beta''$, and ${}_2F_1$ is the hypergeometric function. Technically, however, because the second argument of this function is a complex number it may be more efficient to calculate the spectrum using the time series $P_2^{(n)}(t)$.

The spectrum corresponding to the four-wave mixing directions $2\mathbf{k}_2 - \mathbf{k}_1$ and $2\mathbf{k}_1 - \mathbf{k}_2$ is obtained from Eq. (16) taking $n = -1$ and $n = 2$, respectively, and for $n = 2$ is shown in Fig. 2 as a function of the pulse area. In order to estimate the relation between the decay rate and the nonlinear parameter we have used the expressions following from the microscopic consideration⁷ $\beta' \approx 12\omega_B/\pi$ and $P_{\text{sat}}^2 = 7/32$, where $\hbar\omega_B$ is the exciton Rydberg.

The spectrum has two typical features. The first one consists of branches detached from the exciton frequency with increasing pulse area. These branches for sufficiently high amplitudes of the excitation field may manifest themselves on the spectrum in the form of multiple resonances. The second interesting feature is the oscillatory character of the field dependence of $|P_2^{(2)}(\omega_0; \epsilon)|^2$, which is the FWM response at the exciton frequency.

A. FWM spectrum in the limit of negligible EID

We start the discussion of these features from the simplest case $\beta'' = 0$ (vanishing EID) and high P_{sat} (low saturation regime). The first assumption simplifies the effect of the initial conditions on the polarization dynamics, while the second simplifies the relation between the excitation field and the polarization of the immediate response, so that

$$\tilde{P}_2(t''_+; \kappa) = -i[\epsilon_1 \theta_{10}(\tau) e^{-i\kappa + i\phi_1} + |\epsilon_2| e^{i\phi_2}], \quad (18)$$

where $\phi_1 = \arg[\epsilon_1 \theta_{10}(\tau)]$ and $\phi_2 = \arg(\epsilon_2)$.

It is interesting to note that according to Eqs. (4) and (15) in this approximation since only $\Omega_{\pm 1}(t)$ differ from zero the

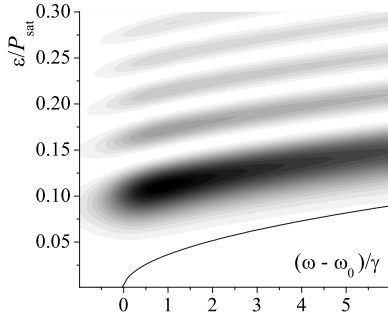


FIG. 3. The form of the four-wave mixing spectrum in low saturation regime in the case of negligible EID ($\beta''=0$) and $\beta'/\gamma = 2.5 \times 10^2$. The solid line shows the position of the resonant frequency determined by $\omega = \omega_0 + \beta I + \eta\gamma$.

dynamics of the polarization is described by the equation of motion for a one-dimensional (1D) tight-binding model with the time-dependent coupling between neighboring sites $\propto \beta |\epsilon_2 \epsilon_1 \theta_{10}(\tau)| e^{-2\gamma t}$. The relation between the amplitudes of the excitation at different sites gives the relation between the amplitudes of the signals corresponding to multiwave mixing. Initially the excitation is localized on sites $n=0, 1$ and with time it propagates along the chain giving

$$P_2^{(n)}(t) = -e^{in(\phi_2 - \phi_1 - \pi/2) + i\phi_2} \exp\left\{-i\omega_0 t - \gamma t - \frac{i\beta I}{2\gamma}(1 - e^{-2\gamma t})\right\} \times \{|\epsilon_1 \theta_{10}(\tau)| J_{n-1}[\eta(1 - e^{-2\gamma t})] - i|\epsilon_2| J_n[\eta(1 - e^{-2\gamma t})]\}, \quad (19)$$

where J_n are the Bessel functions of the first kind, $I = |\epsilon_1 \theta_{10}(\tau)|^2 + |\epsilon_2|^2$, and $\eta = \beta |\epsilon_2 \epsilon_1 \theta_{10}(\tau)| / \gamma$. Deriving Eq. (19) we have used the Jacobi-Anger expansion,²² $\exp(iz \cos \kappa) = \sum_{n=-\infty}^{\infty} i^n J_n(z) e^{in\kappa}$.

The spectrum of the four-wave mixing signal corresponding to $n=2$ is shown in Fig. 3. Shortly after the excitation, for $t \ll 1/2\gamma$, the amplitude of the multiwave mixing signal drops exponentially with the order of mixing, $\sim (\eta\gamma t/2)^n$. The exponential drop holds asymptotically in time if $\eta < 1$. This result agrees with the perturbational approach. The situation, however, drastically changes if the parameters of the system are such that $\eta \gg 1$. In this case, the intensity of the multiwave mixing signal becomes independent of its order starting time $t \gtrsim -\ln(1 - \eta^{-1})/2\gamma \approx 1/2\beta |\epsilon_2 \epsilon_1 \theta_{10}(\tau)|$. This consideration suggests naturally to identify $\eta=1$ as a critical value that separates perturbative and nonperturbative regimes.

The detailed form of the spectrum is essentially determined by the fact that the effective coupling between the modes vanishes with time. Right after arrival of the second pulse the polarization oscillates with the frequency detuned from the exciton frequency by the value $\sim \beta I + \eta\gamma$, where the second term estimates the contribution of the Bessel functions. This detuned frequency qualitatively describes the dependence of the frequencies of the detached resonances on the pulse area (see the bold solid line in Fig. 3). In particular it shows that the resonances in the spectrum widen with increasing nonlinear parameter β . In the opposite limit, t

$\gg 1/\gamma$, the coupling between the modes vanishes and, as a result, the exciton polarization oscillates with the nonmodified exciton frequency ω_0 . These oscillations give rise to the resonant behavior at the exciton frequency, for which dependence on the pulse area is determined by the asymptotic value of the Bessel functions $J(\eta)$. Therefore, the response at the exciton frequency essentially depends on whether the system is in perturbative or nonperturbative regime. We illustrate the difference between these regimes considering the effect of the positive and negative delay times, where the positive delay signal is understood as the signal in the direction determined by the second excitation pulse, i.e., $\mathbf{k}^{(n)} = \mathbf{k}_2 + n\Delta\mathbf{k}$ with $n < 0$. The expression for the negative delay time can be obtained from Eq. (19) by considering the signal in the ‘‘conjugate’’ direction $\bar{n} = 1 - n$ and exchanging $\epsilon_1 \leftrightarrow \epsilon_2$.

In the perturbative regime, $\eta \ll 1$, we obtain

$$|P_2^{(n)}(\omega_0; \tau > 0)|^2 \sim 4\eta_0^{2n-2} |\epsilon_1 n \theta_{10}^{n-1}(\tau)|^2, \\ |P_2^{(n)}(\omega_0; \tau < 0)|^2 \sim 4\eta_0^{2n-2} |\epsilon_1 n \theta_{10}^n(-\tau)|^2, \quad (20)$$

where $\eta_0 = \beta |\epsilon_1 \epsilon_2| / \gamma$. In this regime $|\theta_{10}(\tau)| \approx \exp(-\gamma\tau)$, thus, the positive delay signal decays with time constant $2n\gamma$ while for the negative delay it decays more slowly with the constant $2(n-1)\gamma$. For the case of FWM signal this result agrees with the perturbational calculations.⁹

In the nonperturbative regime the positive and negative delay signals are determined by the oscillating asymptotics of the Bessel functions,²³

$$|P_2^{(n)}(\omega_0; \tau > 0)|^2 \sim \frac{1}{\pi\eta} [|\epsilon_1 \theta_{10}(\tau)|^2 + |\epsilon_2|^2 \\ + (-1)^n (|\epsilon_1 \theta_{10}(\tau)|^2 - |\epsilon_2|^2) \sin(2\eta)], \\ |P_2^{(n)}(\omega_0; \tau < 0)|^2 \sim \frac{1}{\pi\eta} [|\epsilon_1|^2 + |\epsilon_2 \theta_{10}(\tau)|^2 \\ + (-1)^n (|\epsilon_1|^2 - |\epsilon_2 \theta_{10}(\tau)|^2) \sin(2\eta)]. \quad (21)$$

Writing this expression we have neglected the oscillating term $\propto \cos(2\eta)$ vanishing as $(|\epsilon_1 \theta_{10}(\tau)|^2 + |\epsilon_2|^2) / \eta^2$ with increasing pulse area. Thus, for both positive and negative delays the response at the exciton frequency saturates at the oscillations with the period $\eta_T = 2\pi$. The strong asymmetry between these cases specific for the perturbative regime does not hold any longer and the only difference is the phase of the oscillations. It should be noted that in Ref. 24 the saturation of the FWM response found in the numerical calculations was attributed to the renormalization of the pulse area by the EID and EIS. In order to provide the qualitative comparison we calculate the pulse area corresponding to the transition to the nonperturbative regime. Setting $\eta_{cr} \sim 1$ [compare with Fig. 4(d)], assuming no delay between the incoming exciting pulses, $\tau=0$, we obtain

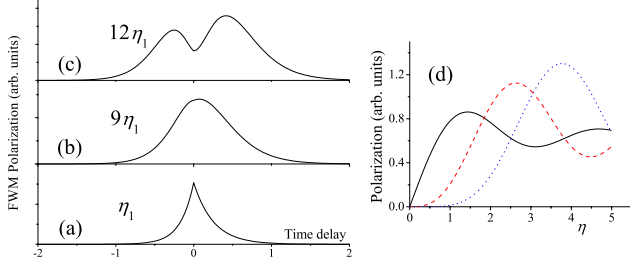


FIG. 4. (Color online) Transition from the perturbative to non-perturbative regimes. Left panel. The dependence of the FWM polarization at the exciton frequency on the delay time for different values of the parameter η : (a) $\eta=0.32$, (b) $\eta=2.88$, and (c) $\eta=3.84$. In order to facilitate comparison with the results obtained in Ref. 9 the graphs are calculated for $\epsilon_2/\epsilon_1=\sqrt{3}$. The low, comparing to Ref. 9, value of η in (c) is due to neglected Pauli blocking in Eq. (19). Right panel (d). Multiwave mixing polarizations, linear-response direction $n=-1$ (solid line), FWM $n=-2$ (dashed line), and six-wave mixing direction $n=-3$ (dotted line) as functions of η .

$$\epsilon_{cr} = \sqrt{\frac{\eta_{cr}\gamma}{\beta}} \sim \sqrt{\frac{\pi\gamma}{12\omega_B}} \approx 0.2, \quad (22)$$

where for the estimate we choose $\gamma/\omega_B \approx 0.1$. Found value of ϵ_{cr} is of the same order of magnitude as that found in Ref. 24 indicating that the saturation effect observed in Ref. 24 is related to the transition to the nonperturbative regime. The present consideration, however, suggests that the origin of the nontrivial dependence of the response on the pulse area is the redistribution of the excitation over multiwave mixing modes. Considering the identity $\sum_n J_n^2(z)=1$ for the limits $z \ll 1$ and $z \gg 1$ it can be seen that such redistribution is especially effective in the nonperturbative regime resulting in essential suppression of the FWM response.

The crossover from perturbative to nonperturbative regimes, as it is seen in the dependence of the FWM polarization on the delay time, is shown in Fig. 4. Similarly to the results obtained in Ref. 9, when the pulse area of the external excitation increases the dependence of the FWM polarization at the exciton frequency on the delay time is no longer exponential. First the shoulder for the positive delay starts to form and then the dependence becomes nonmonotonous. As follows from Eq. (19) for the positive delay time one has

$$\frac{\partial}{\partial \tau} |P^{(-1)}(\omega_0; \tau=0)|^2 \propto -2J_1(\eta) [\eta(|\epsilon_2|^2 - |\epsilon_1|^2)J_0(\eta) + 2|\epsilon_1|^2 J_1(\eta)]. \quad (23)$$

Thus, when $\epsilon_1=\epsilon_2$ the transition from decay to rise at $\tau=0$ does not occur. The dependence, however, becomes flat in this case as η reaches a zero of the Bessel function. The smallest such value is $\eta \approx 3.83$.

As has been discussed above [and is evident from Eqs. (20) and (21)], the transition to the nonperturbative regime results in significantly changed relation between the multiwave mixing polarizations of different orders, which is illustrated by Fig. 4(d). For $\eta \geq 1$ the relation does not follow the exponential law prescribed by the perturbation theory. Because of the nonmonotonous dependence at specific values

of η the magnitude of the six-wave mixing polarization may exceed that of the FWM polarization, which, in turn, may be higher than the polarization in the direction of the linear response.

It should be emphasized that the mechanism of the oscillatory dependence of the response at the exciton frequency is different from the Rabi oscillations,^{25,26} which would correspond to the nonmonotonous dependence of the immediate response on the excitation field. In the case under consideration the oscillations are the result of the free dynamics of the exciton polarization when the external field is turned off. The physics of the Rabi oscillations and of the oscillations of $|P_2^{(n)}(\omega; \eta)|^2$, of course, are essentially the same. As noted above the dynamics of the polarization in the case under consideration appears analogous to a 1D tight-binding model with vanishing coupling between the neighboring sites. In quantum-mechanical terms it can be described as a multiple level system, where the levels correspond to the exciton modes, with time-dependent field $V_{ij}(t)$, which couples different levels. Depending on the “area” of the off-diagonal elements, $\int dt V_{ij}(t)$, one has the oscillations of the final populations of the different levels. Translated to the language of the multimixing signals $P_2^{(n)}$ this result implies the oscillations of $|P_2^{(n)}(\omega_0)|^2$ since asymptotically, as has been noted, one has the dynamics determined by the nonperturbed exciton frequency.

B. Effect of the excitation induced decay

In order to study the effect of the EID on the spectrum (compare Figs. 2 and 3) we use Eq. (17) assuming that $|\epsilon_1 \theta_{10}(\tau)| = |\epsilon_2| = \epsilon$. Considering the asymptotic values of the hypergeometric function²⁷ in the limit $\eta'' = 2\beta'' \epsilon^2 / \gamma \gg 1$ we can approximately present the spectrum in the form

$$P_2^{(n)}(\omega) \approx \frac{\epsilon}{\sqrt{\eta''}} \left[C(\omega; X) A_2 e^{-iX \ln \eta''} + \frac{1}{\eta''} \frac{A_1}{iX - 1/2} \right], \quad (24)$$

where

$$C(\omega; X) = \frac{\Gamma(1/2 - i\omega)\Gamma(1/2 - iX)}{\Gamma(1 - i\omega - iX)} \quad (25)$$

and $A_1 = A_n(1)$, $A_2 = A_n(1/2)$ with $A_n(p)$ depending on X only (i.e., on the relation between EIS and EID),

$$A_n(p) = \frac{1}{2\pi} \int_{-\pi}^{\pi} d\kappa (1 + e^{-i\kappa}) e^{-in\kappa} \times \exp \left\{ - (iX + p) \ln \left[2 \cos^2 \left(\frac{\kappa}{2} \right) \right] \right\}. \quad (26)$$

Similarly to the case of negligible EID, the response oscillates and reaches the saturation in the high excitation limit $\eta'' \gg 1$. At the exciton frequency the magnitude of the signal is

$$|P_2^{(n)}(\omega_0)|^2 \sim \frac{\pi\epsilon^2}{X\eta''} |A_2|^2 \tanh(\pi X). \quad (27)$$

This saturation also is of the dynamical origin since we work in the regime of weak saturation due to the Pauli blocking ($\epsilon/P_{\text{sat}} \ll 1$). The significant difference with the previously considered situation is that now the minima of the spectrum as a function of the pulse area are not equally spaced as the oscillating part has the form $\propto \cos(X \ln \eta'')$. Thus, with the decreasing ratio between the real and imaginary parts of the nonlinear parameter the crossover from linear to logarithmic scale occurs.

Finally, we discuss the effect of the saturation due to the Pauli blocking described by parameter P_{sat} . Qualitatively, this effect can be understood as follows. The nonmonotonous behavior of the semiconductor response on the pulse area studied above is supported by unrestricted increase in the polarization of the immediate response [see Eq. (18)]. However, the saturation effect renormalizes the pulse area so that the magnitude of the polarization cannot exceed P_{sat} . For example, from the perspective of the discussion of the effect of EID this means that Eq. (24) remains valid only if two restrictions are met, $2\beta''\epsilon^2/\gamma \gg 1$ and $\epsilon < P_{\text{sat}}$. This imposes the restriction of the decay rate to be sufficiently small, $\gamma \ll \beta''P_{\text{sat}}^2$. In the similar way the saturation affects the transition to the nonperturbative regime obtained analyzing the semiconductor response in the limit of vanishing EID. With this regard it should be noted that the critical value of the pulse area estimated in Eq. (22) is close to $P_{\text{sat}} = \sqrt{7/32} \approx 0.47$ while remains smaller for low-temperature ratios of the decay rate γ and the exciton binding energy.

III. CONCLUSION

We have studied nonperturbative effects in four-wave mixing spectra of semiconductors. These effects are analyzed

using the exact solution of the nonlinear equation of motion of the exciton polarization taking into account EIS, EID, and the saturation effect phenomenologically. We found that the interaction between the excitons accounted by EIS leads to two specific spectral features—a split of the exciton peak and a nonmonotonous dependence of the response at the exciton frequency ω_0 on the magnitude of the external field. The important characteristic of the splitting is that new spectral features should appear at frequencies higher than ω_0 . This allows one to make a distinction between the effect of interaction of the exciton modes and the manifestation of bound biexciton states, which should modify the spectrum at frequencies lower than ω_0 .

We would like to emphasize that these effects do not appear in any order of the perturbational ($\chi^{(n)}$) approach. It can be shown that the appearance of additional spectral features can be traced as a divergence of the perturbational series. It should be stressed that the crossover from the perturbative to nonperturbative regimes is governed by parameters that essentially depend on the decay rate [e.g., $\beta|\epsilon_1\theta_{10}(\tau)\epsilon_2|/\gamma \sim 1$ in the case $\beta''=0$]. This means that the condition of low excitation itself does not necessarily warrant the validity of the perturbation theory. As an ultimate example one can consider the model with $\gamma=0$ when the spectrum (for $\tau=0$) has the form $P(\omega) \propto 1/\sqrt{(\omega-\omega_0-\beta I)^2-\beta^2 I^2}$ with the exciton peak being split for arbitrary low excitations.

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