RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 49, NUMBER 15 15 APRIL 1994-I

Polarization dependence of dephasing processes: A probe for many-body effects

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(Received 7 February 1994)

The spectra of the diffracted wave in femtosecond-four-wave mixing and their dependence on the polarization of the driving fields are shown to be a very sensitive probe for the fundamental scattering processes that govern the coherent dynamics in a semiconductor. We demonstrate that the total dephasing rate is the result of a delicate balance between self-energies and vertex corrections. Particle interactions of the polarization scattering type are shown to be important contributions to the nonlinear optical response in the coherent regime.

The coherent dynamics of optical excitations in semiconductors is dominated by many-body effects. A number of studies employing transient four-wave mixing (FWM) demonstrated that a basic quantum-mechanical effect, i.e., the Fock exchange between carriers, governs the coherent evolution of the excitation.[1-6] This level of description, however, completely fails to explain the dependence of the FWM signal on the polarization of the driving fields, which has been subject to intense research only recently.[7-10]

For the case of resonant excitation of the exciton, different signal strengths,[10] different decay times,[9] and different temporal signatures[7] have been observed for identical and orthogonal polarization of the two incident beams, respectively. Reference 9 has associated the different decay times to a spatially inhomogeneous coupling between different spin states originating from disorder in quantum-well samples. For ideal semiconductors the excitation-induced dephasing,[10] which is based on excitonic screening, is able to explain the different signal strengths. In this case the polarization dependence is due to the fact that the direct Coulomb-scattering contributions vanish for orthogonal linear polarization, where only the contributions of exchange type remain. A consistent many-body treatment of resonant excitation of the exciton including vertex corrections and corresponding exchange contributions in addition to the excitonically screened Hartree-Fock self-energy has been formulated only for the quasiequilibrium limit,[11] however, is out of reach in the coherent regime.

In this paper experimental data for excitation within the continuum of states are presented. For this case a consistent theoretical description based on carrier-carrier scattering can be worked out. We demonstrate that the polarization dependence of the FWM spectra reflects the different types of many-body processes in a very distinct way.

In the experiment we employ a self-diffraction geometry of transient FWM using transform-limited, tunable, Gaussian optical pulses of 140-fs duration. Spectra of the diffracted wave are taken as a function of time delay between the two incident pulses. The spectral resolution is about 1 meV. We excite in the low density limit (total carrier density $\approx 10^{15}$ cm^{-3}) in order to be within the third-order perturbation regime. More details of the experimental setup can be found in Ref. 6. The two incident beams with wave vectors $\mathbf{q}_1$ and $\mathbf{q}_2$, respectively, are linearly polarized either parallel (colinear) or orthogonal (cross linear). Under these conditions the diffracted wave propagating in the direction $2\mathbf{q}_2-\mathbf{q}_1$ evinces the same polarization as the beam with wave vector $\mathbf{q}_1$. The sample under investigation is a high-quality single crystal of germanium, which is antireflection coated on both sides and optically thin (optical density of 0.65 at the direct gap exciton and the continuum states under investigation). It is immersed in liquid helium at a temperature of 1.8 K. Figure 1 compares experimental spectra for colinear (a) and cross-linear (b) excitation within the continuum of states (7 Ry above the direct gap). The linear absorption spectrum and the laser spectrum are depicted in the background. For the colinear case one narrow line at the exciton and a much broader contribution of the continuum states is observed. At zero time delay the peak height of the exciton is ten times the peak height of the continuum. For later time delays the continuum gains relative strength similar to results that we obtained earlier[6] using circular polarization. This is attributed to the interference and interaction of continuum and exciton polarizations as already described by the semiconductor Bloch equations in connection with a constant, phenomenological dephasing rate.[6] This interference phenomenon has independently been observed in Refs. 5 and 12.

For the cross-linear configuration the spectra are strikingly different [Fig. 1(b)]. Here the peak height of the exciton is only about half the peak height of the continuum contribution (at zero time delay). This unexpectedly different behavior for colinear and cross-linear excitation is explained by the microscopic theory derived below. Clearly, these results cannot be understood when assuming a constant, phenomenological dephasing rate within the semiconductor Bloch equations for the heavy-hole

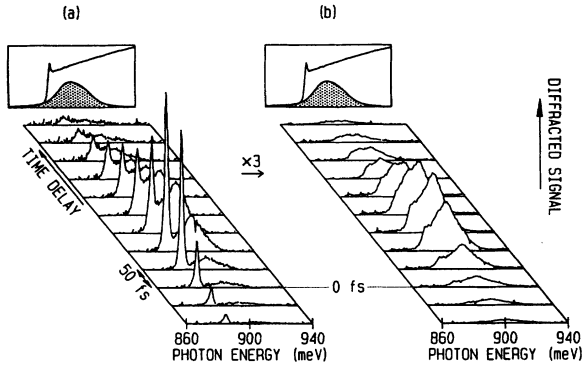

FIG. 1. Experiment: Spectrally resolved femtosecond-four-wave mixing as a function of time delay (in steps of 50 fs). The absorption spectrum and the laser spectrum (shaded) are depicted in the background. Excitation is well (7 Ry) above the direct band gap. Lattice temperature is 1.8 K. (a) Colinear, (b) cross-linear excitation.

and conduction bands only (four-band model). A straightforward calculation shows that this predicts an identical behavior for the two polarization configurations. We also have performed numerical calculations using the semiconductor Bloch equations for the *full* valence-band structure (six-band model). The calculations show that the FWM-signal strength for colinear polarization is somewhat larger than for cross-linear polarization. Importantly, however, the *shape of the spectra remains unaffected* within 10% in striking contrast to the experimental evidence. Any influence from disorder effects[9] can be completely ruled out in the case of the elementary semiconductor germanium. Furthermore we have verified experimentally that the obtained results are independent of the crystal orientation. This rules out linear birefringence and any effects which might arise due to the anisotropic band structure.

A consistent microscopic description of relaxation and dephasing processes can be derived most efficiently by means of nonequilibrium Green's-function techniques.[13] In the scattering contributions of the quantum-kinetic equations all propagators depend on two time variables. This problem has only been treated rigorously for the case of electron-phonon scattering.[14] For the case of carrier-carrier scattering we can employ a Markovian approximation, because here memory effects influence the dephasing only to a minor extent.[15] Anticipating this approximation only equal-time contributions have to be considered. We start from the following general form[14] for the dephasing:

$$\frac{\partial}{\partial t}G^{<}_{cv}(t,t)|_{\text{scatt}}=\sum_{m=c,v}\int_{-\infty}^{t}dt'\{[\Sigma^{>}_{cm}(t,t')G^{<}_{mv}(t',t)-G^{>}_{cm}(t,t')\Sigma^{<}_{mv}(t',t)]+[>\leftrightarrow<]\}\ . \tag{1}$$

The self-energies are the diagonal elements of Σ with respect to the band indices c,v (including spin) for the conduction and (heavy-hole) valence band, respectively, whereas the off-diagonal elements constitute the vertex contributions. From the structure of Eq. (1) it is evident that in a consistent many-body treatment not only the self-energy but also the vertex contributions have to be accounted for. This point has frequently been ignored in the literature. Applying a second-order Born approximation and using a statically screened Coulomb potential w, for example, $\Sigma^{<}_{cv}$ at wave vector $\mathbf{k}$ is given by

$$\Sigma^{<}_{cv}(\mathbf{k},t,t')=\sum_{\mathbf{k}'\mathbf{k}'',nn'=cv}\{w^2(\mathbf{k}')G^{<}_{cv}(\mathbf{k}+\mathbf{k}',t,t')G^{>}_{nn'}(\mathbf{k}'+\mathbf{k}'',t',t)G^{<}_{n'n}(\mathbf{k}'',t,t')-w(\mathbf{k}')w(\mathbf{k}-\mathbf{k}'')G^{<}_{cn}(\mathbf{k}+\mathbf{k}',t,t')G^{>}_{nn'}(\mathbf{k}'+\mathbf{k}'',t',t)G^{<}_{n'v}(\mathbf{k}'',t,t')\}\ . \tag{2}$$

The propagators $G_{nn'}(t,t')$ are factorized into a slowly varying contribution $g_{nn'}(\mathbf{k})$ and a rapidly varying phase factor $\exp[-(\epsilon^{n}_{\mathbf{k}}t-\epsilon^{n'}_{\mathbf{k}}t')]$, which is proportional to the lowest-order solution. $\epsilon^{n}_{\mathbf{k}}$ denotes the single-particle energies within the effective-mass approximation. The time integration in Eq. (1) is performed with respect to the phase factors only, whereas the slowly varying contributions are taken at the upper limit of the integral. This procedure corresponds to the Markovian approximation mentioned above. Applying a similar approach for the kinetic equation of the distribution functions one obtains the usual Boltzmann-type scattering terms. For the dephasing we derive

$$\begin{aligned}\frac{\partial}{\partial t}g^{<}_{cv}(t)|_{\text{scatt}}=-\sum_{\mathbf{k}'\mathbf{k}'',mnn'=cv}\{&[w^2(\mathbf{k}')g^{>}_{cm}(\mathbf{k})g^{<}_{mv}(\mathbf{k}+\mathbf{k}')g^{>}_{nn'}(\mathbf{k}'+\mathbf{k}'')g^{<}_{n'n}(\mathbf{k}'')\\&-w(\mathbf{k}')w(\mathbf{k}-\mathbf{k}'')g^{>}_{cm}(\mathbf{k})g^{<}_{mn}(\mathbf{k}+\mathbf{k}')g^{>}_{nn'}(\mathbf{k}'+\mathbf{k}'')g^{<}_{n'v}(\mathbf{k}'')]s^{mnn'n'}_{-}\\&-[w^2(\mathbf{k}')g^{<}_{mv}(\mathbf{k})g^{>}_{cm}(\mathbf{k}+\mathbf{k}')g^{<}_{nn'}(\mathbf{k}'+\mathbf{k}'')g^{>}_{n'n}(\mathbf{k}'')\\&-w(\mathbf{k}')w(\mathbf{k}-\mathbf{k}'')g^{<}_{mv}(\mathbf{k})g^{>}_{n'm}(\mathbf{k}+\mathbf{k}')g^{<}_{nn'}(\mathbf{k}'+\mathbf{k}'')g^{>}_{cn}(\mathbf{k}'')]s^{mmnn'}_{+}\}+\{>\leftrightarrow<\}\end{aligned} \tag{3}$$

with the definitions $s^{mmnn'}_{\pm}=\mathrm{P}(\pm i/\omega_{mmnn'})+\pi\delta(\omega_{mmnn'})$ (P is the principal value), $\omega_{mmnn'}=\epsilon^{m}_{\mathbf{k}}-\epsilon^{m}_{\mathbf{k}+\mathbf{k}'}+\epsilon^{n}_{\mathbf{k}'+\mathbf{k}''}-\epsilon^{n'}_{\mathbf{k}''}$. The off-diagonal elements of $g_{nn'}(\mathbf{k})$ now contain the phase factors $\exp[-(\epsilon^{n}_{\mathbf{k}}-\epsilon^{n'}_{\mathbf{k}})t]$. It can be seen from Eq. (3) that the random-phase-approximation (RPA) contributions [terms $\sim w^2(\mathbf{k}')$] contain the sum over all possible transitions

and occupations, independent of the subspace of spins in which a certain transition and the dephasing of the polarization takes place. The exchange contributions [terms $\sim w(\mathbf{k}')w(\mathbf{k}-\mathbf{k}'')$], however, only act on the same subspace. This different behavior is the origin of the polarization dependence of the dephasing rate, and hence of the polarization dependence of the FWM signal. In order to see this in detail one has to perform a spatial Fourier analysis with respect to the wave vectors of the involved laser beams[6] and extract the contribution in the direction of the FWM signal. As a result of this we obtain, e.g., for the dephasing contribution of the diffracted signal for colinear polarization in the low density limit $[f_c,(1-f_v)\ll 1]$,

$$\begin{aligned}\frac{\partial}{\partial t}\delta P_{\mathbf{k}}^{2\mathbf{q}_2-\mathbf{q}_1}(t)|_{\text{scatt}} = -\sum_{\mathbf{k}'\mathbf{k}''}\{&[2w^2(\mathbf{k}')-w(\mathbf{k}')w(\mathbf{k}-\mathbf{k}'')][P_{\mathbf{k}}(\delta f^c_{\mathbf{k}'+\mathbf{k}''}-P_{\mathbf{k}'+\mathbf{k}''}\delta P^*_{\mathbf{k}''})s_+^{cccc}-P_{\mathbf{k}+\mathbf{k}'}(\delta f^c_{\mathbf{k}''}-\delta P^*_{\mathbf{k}'+\mathbf{k}''}P_{\mathbf{k}''})s_-^{cccc}]\\ &+2w^2(\mathbf{k}')[P_{\mathbf{k}}(-\delta f^v_{\mathbf{k}''}-\delta P^*_{\mathbf{k}'+\mathbf{k}''}P_{\mathbf{k}''})s_+^{ccvv}-P_{\mathbf{k}+\mathbf{k}'}(-\delta f^v_{\mathbf{k}'+\mathbf{k}''}-P_{\mathbf{k}'+\mathbf{k}''}\delta P^*_{\mathbf{k}''})s_-^{ccvv}]\}\\ &+\{\delta f^c_{\mathbf{k}'+\mathbf{k}''}\leftrightarrow -\delta f^v_{\mathbf{k}''},\delta f^v_{\mathbf{k}'+\mathbf{k}''}\leftrightarrow -\delta f^c_{\mathbf{k}''},\delta P^*_{\mathbf{k}'+\mathbf{k}''}\leftrightarrow\delta P^*_{\mathbf{k}''},P_{\mathbf{k}'+\mathbf{k}''}\leftrightarrow P_{\mathbf{k}''},s^{cccc}\leftrightarrow s^{vvvv},s^{ccvv}\leftrightarrow s^{vvcc}\}\\ &-w(\mathbf{k}')w(\mathbf{k}-\mathbf{k}'')[(-P_{\mathbf{k}+\mathbf{k}'}\delta f^v_{\mathbf{k}'+\mathbf{k}''}-\delta f^c_{\mathbf{k}+\mathbf{k}'}P_{\mathbf{k}'+\mathbf{k}''})s_-^{ccvv}-P_{\mathbf{k}}P_{\mathbf{k}''}(\delta P^*_{\mathbf{k}+\mathbf{k}'}-\delta P^*_{\mathbf{k}'+\mathbf{k}''})s_+^{ccvv}]\\ &-w(\mathbf{k}')w(\mathbf{k}-\mathbf{k}'')[(P_{\mathbf{k}+\mathbf{k}'}\delta f^c_{\mathbf{k}'+\mathbf{k}''}+\delta f^v_{\mathbf{k}+\mathbf{k}'}P_{\mathbf{k}'+\mathbf{k}''})s_+^{vvcc}-P_{\mathbf{k}}P_{\mathbf{k}''}(\delta P^*_{\mathbf{k}+\mathbf{k}'}-\delta P^*_{\mathbf{k}'+\mathbf{k}''})s_-^{vvcc}]\ . \end{aligned}\tag{4}$$

The interband polarizations P and δP refer to the laser beams with wave vectors $\mathbf{q}_2$ and $\mathbf{q}_1$, respectively, and δf is the modulation of the occupation function. The product of the first two sets of square brackets in Eq. (4) corresponds to the RPA and the exchange contribution to the self-energy and vertex corrections within the electron-electron channel. The next line describes the electron-hole scattering. The contributions due to hole-hole and hole-electron scattering are abbreviated in the third line. The remaining terms correspond to diagrams with crossing interaction lines. The factor of 2 at all RPA contributions results from the two different spin states which contribute in the same way. For the case of cross-linear excitation these contributions vanish due to a change in the relative sign of the two spin contributions to δf and δP^*P.

The scattering contributions in Eq. (4) can be separated into the Boltzmann-like terms ($\sim P\delta f$) and the polarization scattering (PS) ($\sim PP\delta P^*$).[16] The Boltzmann-like terms are the physical origin of the so-called excitation-induced dephasing.[10] Here the scattering of an occupied state with a polarization leads to the loss of coherence. In contrast to this, the PS describes the scattering of three coherent polarizations not involving any occupied states. We will argue that the PS is the leading contribution.

Figure 2(b) shows FWM spectra (at zero time delay) obtained from solutions of the semiconductor Bloch equations (containing the usual nonlinear optical terms as phase-space filling and static exchange) combined with the microscopic scattering rate [Eq. (4) and a background dephasing rate]. Parameters correspond to germanium; the excitation conditions are depicted in the center of Fig. 2. The excited carrier density is 0.7×10^{15} cm^{-3}. For the colinear case the exciton peak is much larger than the continuum, whereas both are about equal in the cross-linear configuration. This qualitatively reproduces the main features of the experiment (see Fig. 1, zero time delay) which are again depicted in Fig. 2(a) in order to allow for a direct comparison. Figure 3 illustrates the sensitivity of the FWM spectra on the individual scattering contributions. For reference, the colinear case of Fig. 2(b), which includes all scattering contributions, is reproduced in Fig. 3 (upper curve). Artificially neglecting the PS leads to a qualitatively different scenario (middle curve in Fig. 3). Here the exciton has decreased while the continuum has increased, resulting in identical peak heights. *Hence, the main effect of PS is a transfer of oscillator strength from the continuum to the exciton.* Neglecting all vertex corrections and all exchange contributions (lower curve in Fig. 3) enhances the exciton in the spectra; however, it delivers an absolute signal 500 times

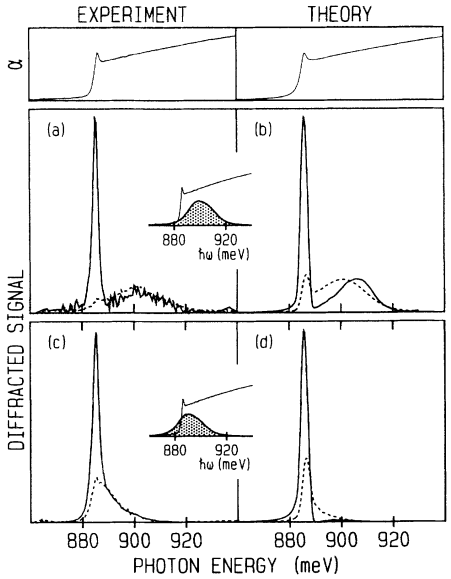

FIG. 2. Spectrally resolved femtosecond-four-wave mixing (at zero time delay). Colinear (solid) and cross-linear (dashed) excitation have to be compared with the experiment. (a) and (b) correspond to the same excitation photon energy (see upper inset) as in Fig. 1, (c) and (d) represent a lower photon energy (see lower inset). For clarity all FWM spectra are normalized according to the continuum contribution. The experimental and theoretical linear absorption (α) spectra, respectively, are shown at the top.

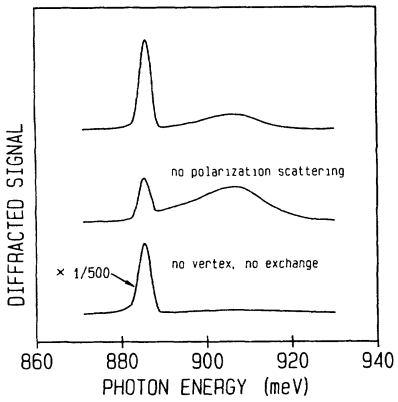

FIG. 3. Theory: Spectrally resolved femtosecond-four-wave mixing (at zero time delay). For reference, the colinear case of Fig. 2(b) including all scattering contributions is reproduced (upper curve). The lower two curves represent two different approximations (see text).

larger. These two *ad hoc* approximations are in striking contrast to the complete theory in which the self-energies are largely compensated by the vertex contributions. In order to further test the theory we vary the excitation photon energy. Respective results are depicted in the lower part of Fig. 2 [(c) experiment, (d) theory]. In the experiment the reduced laser photon energy merges the spectral features of the continuum and exciton, respectively. Again the exciton is more prominent for the colinear as compared to the cross-linear configuration. This is qualitatively described by the theory. In particular the high-energy tail for cross-linear excitation and the dominance of the exciton for colinear polarization are well reproduced. The residual discrepancies (for both excitation photon energies shown) might be due to the Markovian approximation.

In conclusion, the relative coherent contributions of excitons and continuum states, respectively, have been shown to depend dramatically on the polarization of the exciting beams in transient four-wave mixing. A consistent many-body treatment for excitation in the continuum of states, including all self-energies, vertex corrections, and exchange contributions within the second Born approximation is in good agreement with the experimental evidence. We find that the delicate balance between self-energies and vertex corrections governs the dephasing of the optical excitation. The nonlinear optical response in the coherent regime is strongly influenced by polarization scattering.

We acknowledge financial support by the Deutsche Forschungsgemeinschaft. The research of M.W. was supported by the Alfried Krupp-Förderpreis für junge Hochschullehrer of the Krupp-Stiftung. We thank S. W. Koch and R. Binder for valuable discussions on polarization dependences.

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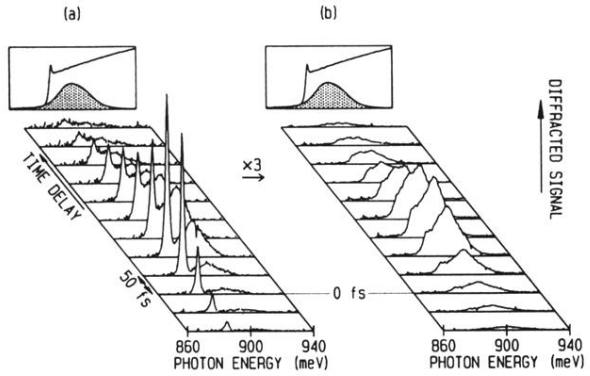

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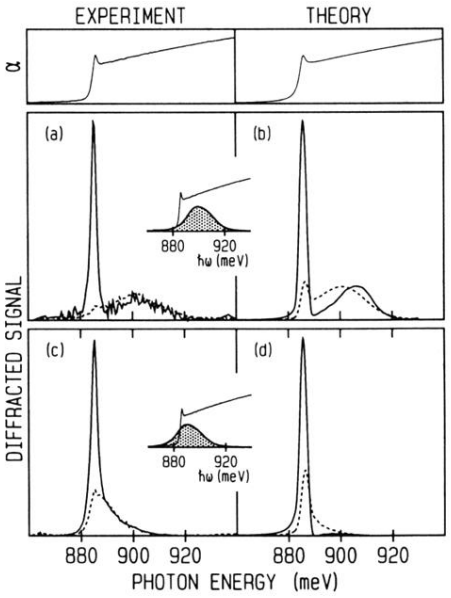

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