Transient Nonlinear Optical Response from Excitation Induced Dephasing in GaAs

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The leading contribution to the polarization dependent four-wave-mixing signal is shown to result from density induced dephasing processes. Experimental observations are in qualitative agreement with theoretical calculations based on the semiconductor Bloch equations where dephasing due to excitonic screening has been taken into account.

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At low excitation densities and low temperatures the nonlinear optical response near the band edge in direct gap semiconductors is dominated by excitonic effects, including Coulomb interactions, exchange effects, and screening [1]. The relative importance of these many-body effects is currently under intense study using ultrafast four-wave-mixing (FWM) techniques [2–5]. The interpretation of most FWM results has been based on purely coherent effects and many-body interactions such as static exchange interaction [6,7]. In materials like GaAs quantum wells or strained bulk GaAs additional information can be extracted from FWM data, if one takes into account the polarization dependence of the twofold degenerate excitons, such as the heavy hole excitons, which include the valence bands with angular momentum states $m_j = \pm \frac{3}{2}$. The optical selection rules for these systems have been thoroughly investigated [8–10]. FWM experiments focusing on optical selection rules have been concerned mainly with the possible spin-flip processes [11–13]. In this Letter we show that polarization dependent FWM signals of initially excited semiconductors are dominated by incoherent many-body effects (dephasing processes). We will focus here on the low density limit where these processes are a consequence of excitonic screening of the carrier-carrier Coulomb potential. In the following we first outline our experimental techniques and results. Then we compare the observations to numerical calculations based on the semiconductor Bloch equations [14–17], in which we also include the contribution describing dephasing due to excitonic screening.

The measurements were carried out at 6 K in a very high quality homogeneously broadened GaAs sample grown by molecular beam epitaxy with a 200 nm GaAs layer sandwiched between $Al_{0.3}Ga_{0.7}As$ layers (necessary to provide support). Uniaxial strain along the growth direction [18,19] lifts the degeneracy between the heavy (hh) and light (lh) hole excitons as shown in the inset of Fig. 1, where the redshift is larger for the lh. The absorption width for the lh exciton is 0.2 meV. The laser pulses for the differential transmission (DT) measurement had an autocorrelation width of 1.5 ps and a spectral width of 4 meV, centered 1 meV below the hh resonance. The probe beam was spectrally resolved with 0.1 meV resolution by an optical multichannel analyzer. The pulses used in the FWM measurement had an autocorrelation width of 3 ps and a spectral width of 0.9 meV to enable the resolution of the hh and lh excitons.

Figure 1 shows a typical DT spectrum of the hh exciton where the pump beam generates an exciton density of $3\times10^{15}/\text{cm}^3$ and is 3 ps advanced with respect to the probe to avoid coherent coupling (a laser spot size of 5×10^{-4} cm^2 is used in estimating the exciton density). The DT spectrum of the hh is nearly symmetric and shows only a negligible shift in the exciton energy (less than 0.1 meV). The change in the oscillator strength is determined by the spectral integral of the hh DT response. The spectrum in Fig. 1 has a net area less than 5% of the positive area, showing that the dominant nonlinear optical response comes from changes in the linewidth (i.e., dephasing rate).

To confirm this, we first assume that the DT spectrum can be taken as the difference of two Lorentzians,

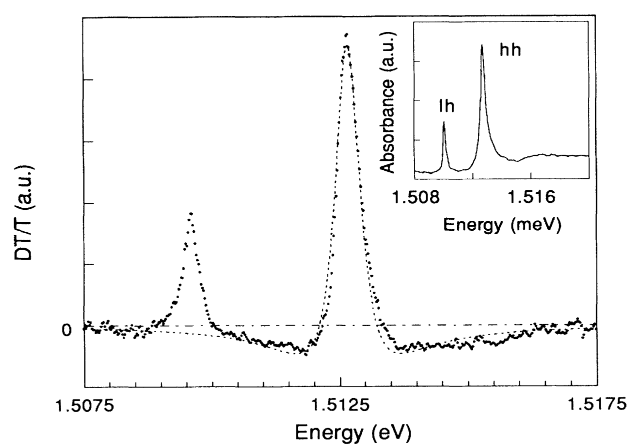

FIG. 1. DT spectrum at an exciton density of $3\times10^{15}/\text{cm}^3$. The dashed line is a fit to the assumed subtracted Lorentzian. Inset: Linear absorption of the sample.

$f[\gamma_2/(\delta^2+\gamma_2^2)-\gamma_1/(\delta^2+\gamma_1^2)]$, where f is the oscillator strength, δ is the detuning from line center, and $\gamma_{1,2}$ are the dephasing rates without and with the pump beam, respectively. From the zero crossings in the DT spectrum occurring at $\delta^2=\gamma_1\gamma_2$, we can determine the density dependent dephasing rate as well as the low excitation density linewidth. The obtained dephasing rate due to excitation induced dephasing (EID) is in general agreement with earlier FWM measurements [20]. We then fitted the hh spectrum in Fig. 1 with no adjustable parameter except the peak height.

To further study excitation induced dephasing, we examine the self-diffracted FWM signal (in the direction of $2\mathbf{k}_2-\mathbf{k}_1$). Figure 2(a) displays the response at the hh resonance where $\mathbf{E}_1$ and $\mathbf{E}_2$ are σ_+ polarized. A prepulse with wave vector $-\mathbf{k}_2$ arriving 3 ps before $\mathbf{E}_1$ is used to generate excitons ($N=2\times10^{15}/\mathrm{cm}^3$) incoherent with those ($N=2\times10^{15}/\mathrm{cm}^3$) generated by $\mathbf{E}_1$ and $\mathbf{E}_2$. The incoherent excitons affect the FWM signal by changing the EID, leading to a large reduction [a factor of 6 in Fig. 2(a)] of the signal strength. From DT data, the dephasing time (T_2) in the absence of the prepulse is estimated to be 1.7 ps. The presence of the prepulse further reduces T_2. Since FWM signals decay as $T_2/2$, the measurement is pulse width limited (FWHM of the signal with no prepulse is 3.1 ps). Hence, the induced change in T_2 is not readily observable in our measurement. As shown in Fig. 2(a), the presence of σ_+ and σ_- polarized prepulses results in nearly the same reduction in the hh signal amplitude, indicating the *spin independent* nature of interactions involved in EID. Spin flipping of excitons is not important in these measurements since the removal of hh and lh degeneracy significantly decreases the exciton spin-flip rate [21]. In separate measurements we have shown that the combined spin and energy relaxation time of the hh exciton is longer than 20 ps. Density dependent exciton dephasing has also been reported in earlier DT and FWM measurements in GaAs [20,22]. The dephasing, however, was attributed to the spin dependent exchange effects [20].

Figure 2(b) displays $I_\parallel/I_\perp$ at the hh resonances as a function of the exciton density generated by a prepulse arriving 20 ps before $\mathbf{E}_2$, where $I_\parallel$ and $I_\perp$ are the FWM signals obtained with $\mathbf{E}_1\parallel\mathbf{E}_2$ and $\mathbf{E}_1\perp\mathbf{E}_2$, respectively ($\mathbf{E}_1$ and $\mathbf{E}_2$ are linearly polarized). At relatively low exciton density, we find $I_\parallel/I_\perp>10$. The ratio decreases with increasing prepulse intensities.

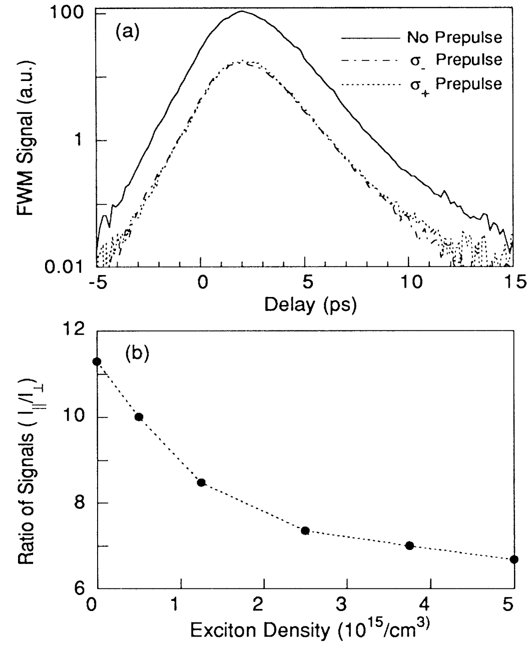

FIG. 2. (a) Effect of σ_+ and σ_- prepulses on the hh FWM response. Measurements as a function of delay between $\mathbf{E}_2$ and $\mathbf{E}_1$. (b) Ratio of co- and cross-linearly polarized FWM signals at the hh resonance as a function of the exciton density generated by the prepulse (the exciton density without the prepulse is $6\times10^{14}/\mathrm{cm}^3$). Dashed lines are to guide the eye.

To analyze the experimental observations we use the semiconductor Bloch equations which describe the optical response of a multiband semiconductor to an external optical field. Within the statically screened Hartree-Fock (SHF) approximation these equations account for phase-space blocking (including band filling and exchange effects) and screening of the Coulomb potential. In this study EID is modeled by the density dependence of the imaginary part of the dynamically screened Hartree-Fock self-energies. We also restrict ourselves to the two optical polarization equations for the $m_j=\pm\frac{3}{2}$ hh excitons because, from the Luttinger-Kane theory, the only consequence of the hh-lh coupling is the mixing of s-like and d-like excitons combined with slight energy shifts, and the creation of d excitons is forbidden in optical transitions.

The equations describing the optical polarization of the two subsystems $s=\{\sigma_+,\sigma_-\}$ are ($\hbar=1$)

$$i\frac{\partial P_s(k)}{\partial t}=[\omega_s(k)-i\gamma_s(k,n)-\Sigma_s^{\mathrm{SX}}(k)-\Sigma^{\mathrm{CH}}(k)]P_s(k)-[1-f_{s,e}(k)-f_{s,h}(k)]\left[\boldsymbol{\mu}_{k,s}\cdot\mathbf{E}(t)+\sum_{k'}W(k-k')P_s(k')\right], \tag{1}$$

where $\omega_s(k)=k^2/2m+E_s$, in which m is the reduced mass and E_g is the unrenormalized band gap. Σ^{SX} and Σ^{CH} are the screened-exchange and the Coulomb-hole self-energy, respectively. W is the screened Coulomb potential and $f(k)$ is the momentum dependent carrier occupation function. The equation for $f(k)$ used in our numerical analysis can be found, e.g., in [15]. Whereas phase-space blocking affects each subsystem, s, according to only the density in that subsystem, the two subsystems are coupled in this model because screening and dephasing depend on the *total exciton density* $n=\sum_s n_s$. The leading contribution of this coupling stems from the density dependent part of the dephasing rate γ. Within the SHF approximation, γ is given by (see, e.g., [1], p. 45)

$$\gamma_s(k,n)=\gamma_0+\sum_{\nu=e,h}\sum_{k'}[f_\nu(k')+g_B(\varepsilon_\nu(k')-\varepsilon_\nu(k))]\,\mathrm{Im}W(k-k',\varepsilon_\nu(k)-\varepsilon_\nu(k'))\,. \tag{2}$$

In quasithermal equilibrium g_B is taken to be the Bose distribution function that describes the thermal occupation of the longitudinal screening quanta that are defined by the imaginary part of the screened Coulomb potential, $\mathrm{Im}W(q,\omega)$. In the following we use a plasmon-pole approximation for excitonic screening (for details see p. 64 of [1]). We fix the limit $q\to\infty$ of the excitonic plasmon dispersion to coincide with the poles of the excitonic polarizability [23], which are approximately at $\omega=E_M(q)=q^2/2M$, where $M=m_h+m_e$ is the total exciton mass. The dispersion of the excitonic plasmon model is then given by $\omega_q^2=(\frac{8}{9}E_B)^2+\omega_{\mathrm{pl}}^2+E_M^2(q)$, where E_B is the exciton Rydberg energy and ω_{pl} denotes the plasma frequency. Because the exciton wave function is centered around $k=0$, we evaluate (2) at $k=0$ and thus obtain a simple analytic expression for the density dependent dephasing.

To describe a two-pulse experiment one has to perform a spatial Fourier analysis of Eq. (1) [7]. This can be done analytically if the density dependence is represented by a Taylor series. Specifically, in the low excitation limit, we keep only the linear term in the expansion of γ:

$$\gamma_s(n)\cong\gamma(n_s^o,n^o)+\gamma'(n_s^o,n^o)\Delta n\,. \tag{3}$$

The superscript "o" denotes the density generated by the prepulse, and γ' is the derivative with respect to n. The coupling of the σ_+ and σ_- subsystems is given by $\gamma'(n_\pm^o,n^o)\Delta n_\mp$, showing that the density dependence of the coupling is essentially given by the *slope* of the density dependent dephasing. More importantly, the EID leads to an *additional* FWM signal through a spatial modulation in the exciton dephasing rate where the strength of the signal is proportional to the square of the slope. An analytic perturbation analysis shows that the two gratings ($\Delta n_\pm$) are out of phase if the pulses are polarized perpendicularly, yielding no dephasing induced contributions. For parallel polarized pulses, the gratings are in phase and yield a nonzero contribution.

A typical variation of the dephasing rate (with $T=20$ K) is shown in the inset of Fig. 3. The corresponding computed signal ratio $I_\parallel/I_\perp$ is shown by the solid curve. In addition to a strong density dependence, we find also a pronounced mass dependence of γ.

Our analysis shows that the dependence of $I_\perp$ on the exciton density is much weaker than its counterpart $I_\parallel$ because, as discussed above, there is essentially no FWM signal due to EID in $I_\perp$. The signal is affected only by the overall change in the dephasing rate. In contrast, in our parameter regime, $I_\parallel$ is a sensitive function of γ', which decreases with increasing exciton densities due to screening of the exciton-exciton interaction. In the high density limit, FWM signals from EID become small with respect to other contributions. Hence, the ratio $I_\parallel/I_\perp$ approaches unity.

We also plot in Fig. 3 as a dashed line $I_\parallel/I_\perp$ for the same parameters, except without the exchange interaction terms in the semicondcutor Bloch equations. In [7] it was shown that the exchange effects dominate the nonlinear optical signal for a two-band semiconductor. In our current model the contributions from both subsystems, σ_+ and σ_-, experience this effect. Only $I_\parallel$, however, is additionally affected by EID signals, which, of course, are also influenced by the exchange effects. The ratio $I_\parallel/I_\perp$ is enhanced without exchange terms, since the EID signal is only indirectly, via the density grating, influenced by the exchange effects, and hence is less reduced when the exchange terms are omitted.

Comparing the experimental results of Fig. 2(b) with the theoretical results shown in Fig. 3, we find good qualitative agreement. We therefore interpret the observed density dependence of $I_\parallel/I_\perp$ as a measure of the incoherent dephasing processes. Indeed, to the best of our knowledge, no other experiments have been able to clearly separate spin dependent exchange effects from spin independent screening effects. It is interesting to note that the density dependent dephasing rates used to obtain the fit in Fig. 1 deviate from a linear dependence on the exciton density. Using this to obtain γ' we compute $I_\parallel/I_\perp$ and find qualitative agreement with Fig. 2(b) and the solid line in Fig. 3. Further investigations of excitonic

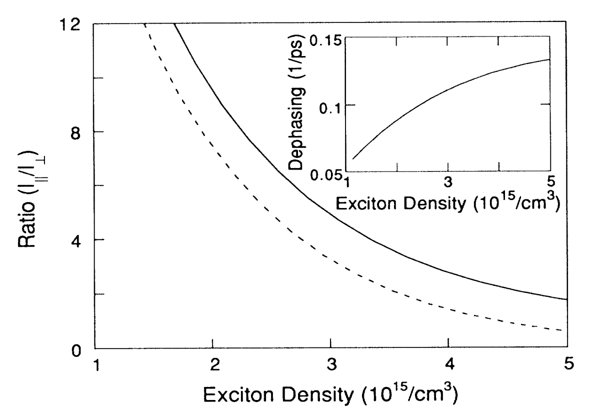

FIG. 3. Theoretical results of the ratio of co- and cross-linearly polarized hh FWM signals as a function of the exciton density generated by the prepulse. Dashed line is the result (divided by 10) without including exchange interactions. Inset shows the extra dephasing induced by the exciton-exciton interaction.

screening processes, along the lines of Refs. [24,25], are expected to result in improved quantitative agreement.

It should be pointed out that because of the removal of the hh-lh degeneracy, the lh continuum is much closer to the hh exciton resonance than in an unstrained sample. Small numbers of free carriers from the lh transition are generated in DT and FWM measurements. Although the density of carriers is estimated to be less than 10% that of the hh excitons, their presence complicates the interpretation of the experimental result since free carriers are much more efficient in screening the Coulomb interaction than excitons. In fact, at an excitation level $\sim 10^{14}$ cm^{-3}, the ratio $I_{\parallel}/I_{\perp}$ increases if the pulses are tuned above the hh resonance. The ratio, however, remains nearly constant when the pulses are tuned at or below the hh resonance, indicating that contributions from free carrier screening are not significant under these conditions.

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