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Polarization-dependent picosecond excitonic nonlinearities and the complexities of disorder

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Time-resolved emission of the hh1 exciton transient four-wave-mixing response in multiple quantum wells shows the unexpected result that the signal emission is either a photon echo or a free polarization decay, depending on the relative polarization of the excitation beams. The energy and temperature dependences of the dephasing rates and the transport and saturation characteristics show that the two excitations are significantly different. Explanations for the disappearance of the echo, which arises due to the presence of disorder, and the origin of the prompt signal are considered.

Picosecond optical excitation of excitons in semiconductors leads to coherent nonlinear optical effects such as photon echoes, free polarization decay, and quantum beat phenomena. The physics underlying these interactions was first described in simple systems based on the optical Bloch equations (OBE). In semiconductors the description is complicated by the Coulomb interaction and the fermionic nature of the carriers. However, progress in the development of a parallel treatment of optical interactions in semiconductors resulting in a set of semiconductor OBE (Ref. 1) has greatly facilitated understanding. Work in this area is the subject of numerous recent studies, not only because this is a challenging many-body problem important for applications to optoelectronic devices, but also because nonlinear spectroscopy is a powerful means of studying materials.

The nonlinear response and excitation dynamics in semiconductor heterostructures, however, is qualitatively changed due to the presence of disorder such as monolayer fluctuations at the $GaAs/Al_xGa_{1-x}As$ interface. Early measurements demonstrated that the low temperature exciton line width in GaAs multiple quantum wells (MQW) is inhomogeneously broadened by the corresponding confinement potential fluctuations.[2] It was initially believed that there were large regions at the interface that were atomically flat. More recent studies have shown that the disorder is due to the presence of monolayer flat islands with a spatial extent of 50–100 Å (Ref. 3) suggesting the existence of at least two scale lengths for interface roughness.[4,5]

Nonlinear optical measurements of excitons in MQW at low temperature have improved the understanding. Transient four-wave mixing (TFWM) showed that in MQW the emission is a photon echo, confirming the resonance is inhomogeneously broadened.[6] Measurements on a single QW with a much narrower hh1 absorption line demonstrated that the resonance was homogeneously broadened.[7] In MQW TFWM measurements of the diffusion coefficient[8] showed a strong dependence on photon energy. The diffusion rate increased through the absorption line center which is evidence for the exciton mobility edge. Theoretical[9] and experimental[10,11] work showed that exciton relaxation ($T < 15$ K) proceeds by phonon-assisted migration between localization sites. In this paper, we report unexpected features regarding the polarization dependence of the time-resolved emission in TFWM. The results cannot be explained based on existing theoretical models and show the importance of including the effects of polarization-dependent interactions as well as the effects of disorder.

Based on the solution of the ordinary OBE, it is well known that for a single resonance the emission in three pulse TFWM [Fig. 1(a), left-hand inset] is a free polarization decay (FPD) emitted coincident with the third pulse.[12] If the system is inhomogeneously broadened, the FPD from the different resonant groups destructively interfere and the prompt coherent emission vanishes. However, at a time following the third pulse given by the time difference between the first two pulses, the interference becomes constructive, leading to an emitted signal (a stimulated photon echo, SPE) for a duration determined

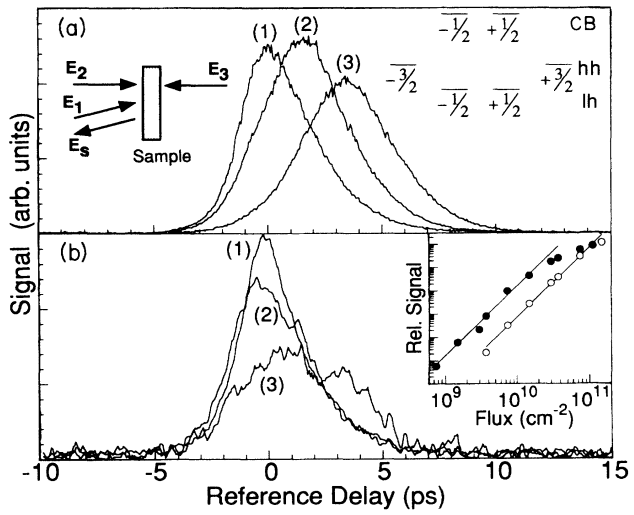

FIG. 1. Time-resolved signal for (a) all fields copolarized and for (b) $E_1 \perp E_2 \| E_3$, $E_s \| E_1$. Delay between E_1 and E_2 is 0 ps for (1), 2 ps for (2), 4 ps for (3) in (a) and (b); all are taken 0.4 meV below the absorption line center. Left inset in (a) is experimental geometry. Subscript on fields also correspond to time ordering. Right inset in (a) shows magnetic substate structure. Inset in (b) shows dependence of signal strength on incident flux (photons/cm^2), filled circles are for $E_1 \| E_2$, open are for $E_1 \perp E_2$, straight lines indicate cubic behavior.

by the inverse inhomogeneous width.

Recently however, it has been shown theoretically that this simple picture based on noninteracting two level systems is not adequate for describing the temporal structure of coherent radiation produced in TFWM resonant with excitons in an ideal semiconductor. Lindberg, Binder, and Koch with similar results by Schäfer, Jahnke, and Schmitt-Rink[13] show that even for resonant excitation of a homogeneously broadened exciton, the time-dependent emission in TFWM can be complex due to exciton-exciton interactions and contributions from the continuum states, leading to additional temporal structure and delay in the emission.

Measurements presented in this paper were obtained on two samples grown by molecular-beam epitaxy and gave qualitatively similar results. The first sample consisted of 65 periods of 96-Å GaAs wells and 98-Å $Al_{0.3}Ga_{0.7}As$ barriers. The hh1 absorption width was 2.2 meV (1.2 meV luminescence Stokes shift). The second sample, with only 10 periods, had an absorption line width less than 1.0 meV (luminescence Stokes shift <0.2 meV). Energy-resolved measurements were obtained using the first sample since the narrow absorption resonance of the higher quality sample was on the order of the inverse pulse width necessary to complete the measurements. The TFWM signal was time resolved by the usual cross-correlation technique. Unless otherwise stated, all measurements were performed at 5.5 K and the autocorrelation width of the incident pulses was 3 ps.

As we discuss in more detail below, it is expected that other than rotating the polarization of the signal, no difference in the FWM response is expected for either all excitation beams linearly copolarized or for $\mathbf{E}_1 \perp \mathbf{E}_2 \| \mathbf{E}_3$, $\mathbf{E}_2 \| \mathbf{E}_1$. Figure 1 shows the surprising result that in the first case the emission is delayed with respect to the third field given by the time between E_1 and E_2 (as shown earlier[11]) while in the second case ($\mathbf{E}_1 \perp \mathbf{E}_2$ excitation) the signal is prompt (i.e., independent of time delay) with respect to the third pulse. [An additional delayed signal is also evident in Fig. 1(b) arising from a strong resonance with a long dephasing time and Stokes shifted by 2.5 meV, most likely due to an impurity bound exciton.] Both signals are *strongly resonant* with the hh1 exciton, approximately centered at the hh1 absorption line center (after corrections for absorption and wavelength-dependent coherence decay rates). Furthermore, we note that at low excitation density, there is no combination of circularly polarized input beams (the natural polarization for this discussion) which gives rise to the rapid dephasing associated with the prompt signal (see below). In the following, we present data which further distinguishes these two signals as well as providing insight into possible explanations.

The exponential decrease in the peak signal strength as a function delay, τ, between E_1 and E_2 is determined by dephasing.[12] Experiments show both signals in Fig. 1 decay monoexponentially, though the dephasing rate in the delayed signal is much slower than for the prompt signal. (The increase in dephasing rate for orthogonal excitation has been reported earlier.[14]) In addition, the dephasing rates differ considerably in energy and temperature dependence. Since the emission delay in Fig. 1(a) varies linearly with the delay τ, we assume the dependence on dephasing is given by $\exp(-4\Gamma_{ph}\tau)$ whereas for the signal in Fig. 1(b), the dependence is given by $\exp(-2\Gamma_{ph}\tau)$.[12] Figures 2(a) and 2(b) show the corresponding energy[15] and temperature dependence (1 meV below line center).

The linear dependence of signal emission on τ coupled with the long dephasing time (in agreement with earlier spectral hole burning measurements[10]) shows that the signal in Fig. 1(a) is a classical stimulated photon echo arising from localized excitons. This is further supported by Fig. 2(a) showing the increase in dephasing rate through line center as demonstrated earlier for localized excitons[8] as well as the fact that the temperature dependence [Fig. 2(b)] follows that due to photon-assisted migration and thermal activation (dashed line) for localized states.[10,16] As seen in the upper inset of Fig. 1(b), this signal also saturates at low intensity ($\sim 5\times 10^8$ excitons/cm^{-2} assuming the absorption is due to the resonance associated with the SPE) due to the finite number of localization sites. In addition the observed exact correspondence between the time delay in the signal and the time τ is not expected at low excitation for homogeneously broadened excitons.[13]

In comparison with the delayed signal, we see that the dephasing rate for the prompt signal is larger and nearly energy independent [Fig. 2(a)]. In addition, the temperature dependence of the dephasing rate for the prompt signal [Fig. 2(b)] is linear and the magnitude and temperature dependence is in agreement with the earlier work discussed above in a homogeneously broadened single QW (Ref. 7) (where the hh1 absorption linewidth was 0.4 meV, in agreement with the reported dephasing rate). In those experiments, it was believed that the exciton was described by an extended state and dephasing was due to single photon scattering. We also note that while the amplitude of the delayed signal saturates at low intensity, the onset of saturation for the prompt signal occurs at a much higher intensity. At high intensities (10^{11} photons/cm^2/sec), the delayed signal saturates, and the prompt signal becomes observable under copolarization excitation. In

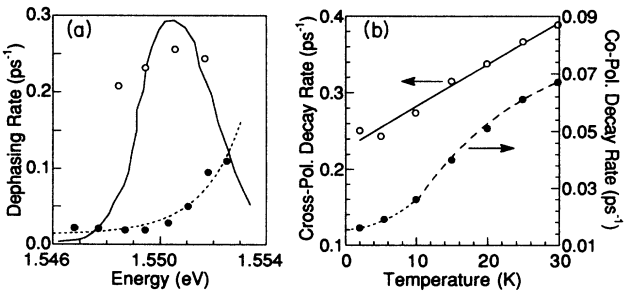

FIG. 2. (a) Energy dependence of dephasing rates for copolarized fields (filled circles, dotted line to guide the eye) and for $\mathbf{E}_1 \perp \mathbf{E}_2$ (open circles). The signal strength is clearly decreasing away from the line center. Solid line is linear absorption. (b) Temperature dependence of both signals 1.0 meV below absorption line center, note difference in scales associated with each. Solid lines are fit to single phonon scattering, dashed lines are fit to phonon assisted migration ($T < 10$ K), and thermal activation ($T > 10$ K).

contrast in the absence of disorder, Lindberg, Binder, and Koch[13] shows the presence of the (second) delayed contribution to the response increases with respect to the first peak with increasing excitation density rather than decreasing.

It is important to note that after accounting for the finite pulse width in the current experiments, the spectral width of the nonlinear response from the prompt signal is considerably narrower than that associated with the echo.[17] The temporal width of the echo is also in agreement with that expected from the linear absorption linewidth.[11]

Finally, we determined the mobility associated with the echo and prompt signal by transient grating experiments.[8] As expected, the diffusion coefficient D, varied between samples. We found that D increases through line center for the echo signal, in agreement with earlier work on localized excitons.[8] However, we found that the D (10.1 and 14 cm^2/sec for the two samples examined) associated with the prompt signal was independent of the excitation wavelength over the resonance and was considerably larger than the D for localized excitons (see Fig. 3). The ratio of the D(prompt)/D(delayed) below line center for the two samples was 2 and 3.5.

In the discussion of these unexpected results, we note that in general the polarization dependence of the FWM signal depends on the details of the symmetry of the electronic excitation. Quantum confinement lifts the valence-band degeneracy at $k=0$ resulting in hh-lh splitting. However, if the energy-level diagram represented in Fig. 1 is a good approximation, it is easy to determine what is expected. The conduction band is $m=\pm\frac{1}{2}$ character and the valence band is $m=\pm\frac{3}{2}$. The axis of quantization and the incident k vectors are perpendicular to the plane of the layers. The dipole selection rules are $\Delta m=\pm 1$ with equal transition moments. For $\mathbf{E}_1\|\mathbf{E}_2$, a population grating (for each frequency group) is induced (E_3 scatters off this grating giving rise to the signal). In the absence of optical alignment and orientation effects[18] the two transitions ($-\frac{3}{2}\rightarrow-\frac{1}{2}$ and $+\frac{3}{2}\rightarrow+\frac{1}{2}$) are independent and at a given frequency each has its own spatial grating. This is also true for $\mathbf{E}_1\perp\mathbf{E}_2$; however, the gratings for the two transitions are spatially out of phase. Hence, although the polarization of the incident fields determines the polarization of the signal, the temporal dependence is not expected to be affected. Note that if the scattering from each grating does not depend on the polarization of E_3, then the signal vanishes for $\mathbf{E}_1\perp\mathbf{E}_2$.

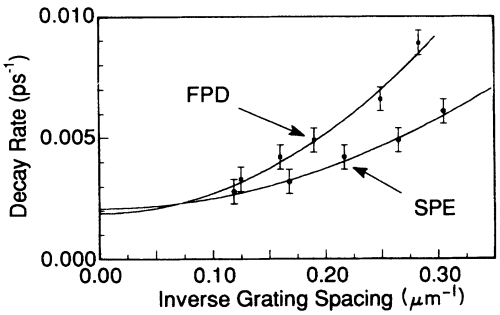

FIG. 3. Grating relaxation rate at absorption line center for SPE and FPD as a function of inverse grating spacing.

Current measurements provide an indication of the mechanism leading to the suppression of the echo for $\mathbf{E}_1\perp\mathbf{E}_2$. Complete randomization of the gratings (corresponding to $\Delta m=+1$ or -1 transitions) on the time scale of the excitation pulse would explain the absence of the echo as would the possibility that the scattering of $\mathbf{E}_3$ from the gratings is polarization independent. Randomization occurs due to spin relaxation but we have shown that this occurs on the time scale of 40 ps, in agreement with earlier measurements,[19,20] much longer than the pulse width. However, our measurements show the unexpected result that corotating circularly polarized $\mathbf{E}_1$ and $\mathbf{E}_2$ with a linearly polarized $\mathbf{E}_3$ gives a *linearly* polarized echo, i.e., the grating induced by the circularly polarized $\mathbf{E}_1$ and $\mathbf{E}_2$ scatters left and right circularly polarized light with equal efficiency. The absence of the echo in the cross-polarized experiments above is thus due to the destructive interference between the two third-order-induced polarizations. The linearly polarized echo observed using circularly polarized $\mathbf{E}_1$ and $\mathbf{E}_2$ shows that both left and right circularly polarized fields couple with the induced grating. The existence of coupling between oppositely rotating fields has been detected in transient absorption experiments using circularly polarized fields reported in earlier work[20] and also observed in our laboratory. The observed coupling is much stronger than expected and is the subject of current studies.[21] However, the coupling may be the result of localization enhanced exciton-exciton interactions.

A detailed understanding of the origin of the prompt signal is incomplete. However, the data shows the response is resonant with the hh1 exciton though the physical properties of this response are distinct from those of the echo. One explanation is that the signal arises from extended excitons. We note that the prompt signal shows no evidence of saturation whereas the echo saturates due to the finite number of localization sites. For an ideal QW, the saturation intensity is over an order of magnitude higher than the range of our measurements.[22] The large wavelength independent dephasing rate is also expected for an extended state exciton where dephasing is due to single phonon scattering. Indeed, both the magnitude and temperature dependence are in agreement with earlier work on a single QW where it is believed that the excitons are extended.[7] And finally as would be expected, the diffusion coefficient associated with this excitation is greater than the diffusion coefficient for the localized exciton, although, the sample-dependent nature of this number shows that this state still experiences scattering from the potential fluctuations due to interface roughness. The simultaneous presence of localized and extended states is not expected in the theory of localization[23] though more recent work challenges this thinking.[24] Alternatively, the states associated with the prompt signal may not be truly extended but confined to large regions of the QW characterized by reduced interface fluctuations whereas the echo originates in areas characterized by increased fluctuations leading to strong localization, a picture consistent with the concept of a bimodal distribution of interface roughness.[4,5]

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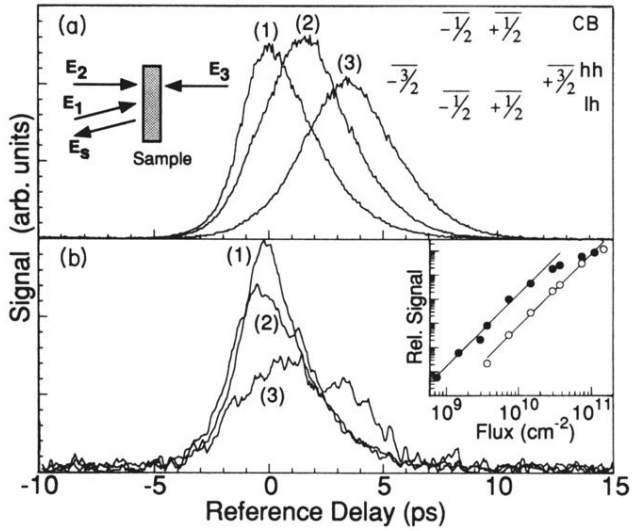

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