Nonlinear optical absorption and dynamics in quantum wells

Min Jiang, Hailin Wang, and Duncan G. Steel
Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan 48109-1120

(Received 24 April 1992; accepted for publication 6 July 1992)

We present measurements of differential transmission and four-wave mixing in GaAs quantum well structures at 1.8 K near the inhomogeneously broadened lowest heavy-hole ($hh1$) exciton resonance using narrow band cw excitation. The data show an *increase* in absorption and an excitation lifetime of order 1–10 μs outside the spectral hole produced by the pump. The long lifetime and the experimentally determined absence of excitation spatial diffusion in this region suggests that optical absorption produces electron-hole pairs that are correlated but separately localized due to disorder. A phenomenological model is proposed to explain the nonlinear response based on two-photon absorption.

The optical properties immediately below the band edge of direct band-gap semiconductors are dominated by excitonic effects. In a quantum well (QW), the strong transient nonlinear optical response associated with the exciton resonance has been shown by numerous theoretical and experimental studies to be due to many-body effects including phase space filling, exchange effects and to a lesser degree, screening.[1] However, the nonlinear response and exciton dynamics are greatly complicated and qualitatively changed by the presence of interface disorder in QW structures.[2] Early measurements suggested large atomically flat areas at the interface.[3] More recent measurements show the presence of monolayer flat island formation on a scale of 50 Å,[4] leading to the proposal that there is a bimodal distribution for island size.[5] At low temperature, excitons can be localized by the interface disorder which leads to strong inhomogeneous broadening of the optical absorption spectrum. Localized excitons, however, can migrate between localization sites by emitting and absorbing acoustic phonons.[6,7]

To improve the understanding of the intrinsic nonlinear optical response and the effects of disorder in GaAs quantum well structures, we present what we believe are the first measurements of the *cw nonlinear response* near the lowest heavy-hole ($hh1$) exciton at low temperature (1.8–5 K) where the effects of disorder is a dominating factor. The experiments are based on *nondegenerate* differential transmission (DT) and four-wave mixing (FWM). Nondegenerate DT measures the sign and magnitude of the imaginary part of the third-order susceptibility while nondegenerate FWM measures the magnitude (squared) of the third-order susceptibility and is useful for determining various relaxation rates.[8,9] While the results of many-body theory[1] have been highly successful in accounting for the nonlinear response observed using short pulse excitation at high excitation density ($>10^9$ excitons/cm^2/layer),[10] we show in this letter that the present understanding cannot account for the experimental results observed at low excitation density under cw excitation. We believe the discrepancy is due to the presence of disorder and propose a possible phenomenological model to explain the data.

The data reported in this letter are obtained in QW structures consisting of 65 periods of 96 Å GaAs wells and 98 Å $Al_{0.3}Ga_{0.7}As$ barriers, grown at 630 °C by molecular beam epitaxy on semi-insulating (100) GaAs substrates with interrupted growth. The $hh1$ exciton absorption linewidth is 2.3 meV [Fig. 1(a)] with a Stokes shift of 1 meV between the $hh1$ exciton absorption and emission. Similar experiment results have also been obtained on several $GaAs/Al_{0.3}Ga_{0.7}As$ samples differing in the number of layers and where the absorption line widths varied from 1.0 to 2.5 meV with a corresponding Stokes shift varying from <0.2 to 1.5 meV. Samples are mounted on a sapphire disk (c-axis normal) with the substrate removed and placed in a liquid helium immersion cryostat.

Measurements are performed using two frequency stabilized cw dye layers. For DT measurements, one laser supplies a pump beam at a fixed frequency Ω_1 while the second laser supplies a probe beam at Ω_2. Both beams are linearly and orthogonally polarized to avoid coherent effects. The nonlinear spectral response proportional to $\mathrm{Im}\chi^{(3)}$ is obtained by measuring the probe transmission as a function of Ω_2 where $\chi^{(3)}$ is the third-order susceptibility. For nondegenerate backward FWM, two nearly degenerate co-polarized beams $\mathbf{E}_1(\mathbf{k}_1,\Omega_1)$ and $\mathbf{E}_1'(\mathbf{k}_1',\Omega_1+\delta)$ with $\delta \ll \Omega_1$ intersect in the sample with a small angle θ between the beams producing a traveling-wave modulation of the absorption and dispersion with a period $\Lambda=\lambda/(2\sin\theta/2)$. The grating is probed by an orthogonally polarized beam $E_2(\mathbf{k}_2,\Omega_2)$ where $\mathbf{k}_2=-\mathbf{k}_1$ producing a coherent signal, proportional to $|\chi^{(3)}|^2$, propagating in the direction $-\mathbf{k}_1'$. Tuning Ω_2 again measures the spectral response.[9] Tuning δ provides information on the excitation decay dynamics probed at Ω_2.[9] This decay rate depends on γ, Γ_{sd}, and Γ_d where γ is the recombination rate, Γ_{sd} is the spectral diffusion rate and $\Gamma_d=4\pi^2D/\Lambda^2$ is the rate due to spatial diffusion where D is the diffusion coefficient.

Figure 1 shows a comparison between the FWM spectrum [Fig. 1(b)] and the DT spectrum [Fig. 1(c)] obtained by tuning Ω_2 [Fig. 1(b) is similar to that reported earlier[7,11]]. Ω_1 is given by the arrow in the $hh1$ linear absorption feature shown in Fig. 1(a). The sharp resonance at $\Omega_2=\Omega_1$ in Fig. 1(b) is the result of spectral hole burning of the inhomogeneously broadened localized excitons. The width of the hole is twice the homogeneous width.[8,9] The comparable DT spectrum [Fig. 1(c)], obtained at the same

 0003-6951/92/361301-03$03.00

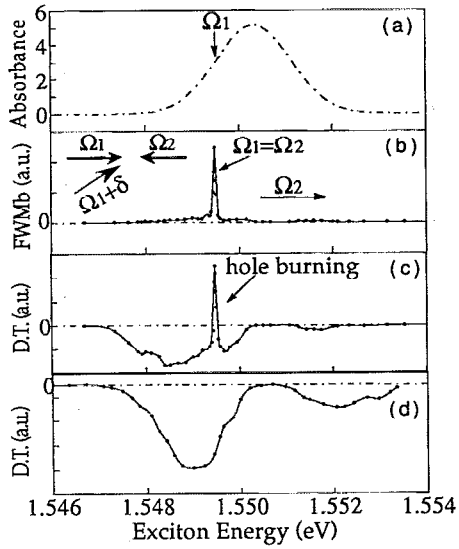

FIG. 1. A comparison of the cw four-wave mixing and differential transmission spectra. (a) The *hh*1 linear absorption spectrum. The arrow shows the location of Ω_1 for (b) and (c). (b) The four-wave mixing spectrum obtained by tuning Ω_2. (c) The corresponding differential transmission spectrum. (d) The differential transmission spectrum obtained when Ω_1 is set to 1.2 eV above the *hh*1 absorption peak.

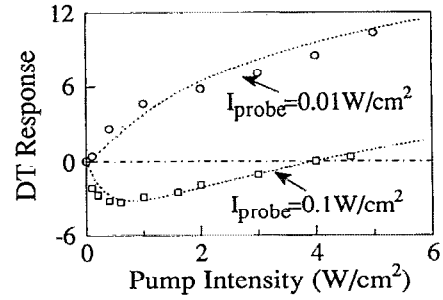

FIG. 2. The intensity dependence of DT spectra as a function of pump intensity for two different probe intensities. Measurements are made for the case of Fig. 1(c) where Ω_1 is set 1.1 meV below Ω_1. The dotted curves show the fitting based on a two component model: an incoherent two photon stepwise excitation dominates at high probe intensity (square) and a single saturation type nonlinear response dominates at low probe intensity (circle) characterized by a smaller saturation intensity than that of a two photon-type nonlinear response.

excitation intensity ($\sim$0.5 W/cm^2), shows the sharp resonance again corresponding to the spectral hole seen in Fig. 1(b). As expected from phase space filling effects, a decrease in absorption is observed in the spectral hole (a positive signal corresponds to a *decrease* in absorption). However, away from the spectral hole, the DT measurement shows an unexpected *increase in absorption*. It is easy to see that the DT response ($\propto \mathrm{Im}\chi^{(3)}$) away from the spectral hole is much larger than expected based on FWM ($\propto |\chi^{(3)}|^2$). Figure 1(d) shows the DT spectrum obtained when the pump frequency Ω_1 is tuned to 1.2 meV above the *hh*1 absorption line center where no spectral hole is expected or observed.[12] This spectrum [Fig. 1(d)] demonstrates that the increase in absorption around *hh*1 is *not related to the spectral hole*. A similar DT spectrum is observed when Ω_1 is tuned above the band edge where free carriers are directly excited. In contrast no signal is obtained when Ω_1 is tuned well below the *hh*1 resonance.

Measurements of the intensity dependence of the FWM signal show that the spectral hole amplitude depends linearly on each of the intensities of the three input beams, indicating that there is minimal contribution from higher order terms in the susceptibility. In the usual approach of estimating the exciton density based on a radiative lifetime of order 1 ns and the input intensity, the exciton density is of order 10^7–10^8 excitons/cm^2/layer. However, even at this low excitation level, the DT signal in Fig. 1(d) is not linear in the pump intensity. In fact, the response also *depends on the probe intensity*. Figure 2 shows the unusual intensity dependence of the DT response as a function of pump intensity for two different probe intensities. Measurements are made for the case of Fig. 1(c) where Ω_2 is set 1.1 meV below Ω_1.

We further characterize the nonlinear response by measuring the relaxation time in the region showing increased absorption. Independent measurements using amplitude modulation DT spectroscopy[13] and FWM by tuning δ[7] show a decay time of order 1–10 μs in contrast to the 0.5–1 ns excitonic recombination rate obtained in this sample by time-resolved luminescence and FWM at the hole burning resonance. While the slow time scale suggests that the nonlinear response could arise due to photorefractive or thermal effects, the first possibility is eliminated since we have determined that there is no energy transfer between beams as would be expected in the presence of two beam coupling. Thermal effects are also eliminated for two reasons: (1) The DT signal strength does not decrease when the sample is immersed in liquid helium as would be expected since in liquid helium, the induced temperature gradient is dramatically reduced; (2) more importantly, the absence of any dependence of the grating decay on the grating spacing sets the upper limit of the diffusion coefficient at 3×10^{-4} cm^2/s. If the nonlinear response were due to thermal effects, then this measurement would correspond to a thermal conductivity at least 4 orders or magnitude below that of GaAs. Hence, we conclude the excitation is electronic in nature and note that the long lifetime results in an estimated excitation density four orders of magnitude higher than that expected based on a 0.5–1.0 ns exciton lifetime.

In discussing these results we first note that many-body effects such as exciton-exciton interactions, band-gap renormalization, and screening can produce a shift or a broadening of the resonance, leading to regions of increased absorption. However, these features resemble the first or second derivative of the resonance, clearly not in agreement with the measurement. Hence, while the current theory has been highly successful in interpreting experimental results obtained on short time scales at high excitation density,[1,10] the results show that the theory does not describe the leading terms in the low intensity cw nonlinear response measured by differential transmission. We would like to stress that identical behavior has been observed in the three different samples we investigated. Earlier experimental evidence for this effect was reported in transient

DT measurements using picosecond lasers.[2] At zero time delay, the low-temperature excitonic response showed a decrease in absorption due to bleaching, yet on longer time scales ($\sim$100 ps), the DT signal changed sign, showing an increase in absorption.

The nonlinear response outside the spectral hole can be qualitatively explained by a phenomenological model involving an incoherent two photon stepwise excitation along with an ordinary saturating type nonlinear response (not associated with the spectral hole). In a rate equation description for this system [i.e., a simple two level system (2LS) and an independent three level system (3LS)], a probe dependent DT response similar to the data is obtained as shown by the dotted lines in Fig. 2. In this model, the transition rates for both transitions in the 3LS are comparable. When the pump beam is resonant with the transition from level 1 to level 2 and the probe beam is resonant with the transition from level 2 to level 3, the DT signal due to the 3LS would show an increase in absorption if the beam intensities are reasonably low. However, if the 2LS saturation intensity is smaller than saturation intensities for the 3LS, at very low intensities, the DT signal is dominated by the 2LS. Hence, decreased absorption is observed as shown in Fig. 2. As the probe intensity increases, the relative importance of the 3LS will increase due to the small saturation intensity for the 2LS, resulting in increased absorption in the DT response. Finally, when the pump beam intensity is very high such that both transitions in the 3LS are saturated, the stepwise two-photon transition will be overwhelmed by the saturation effect and an overall decrease in absorption will be observed as shown in Fig. 2.

The relaxation measurements also provide some additional insight into the microscopic origin of the observed nonlinear response in the QW. In particular, even though the nonlinear response is clearly associated with the *hh*1 exciton, the long lifetime of the excitation is not consistent with the presence of ordinary excitons, nor is the negligibly small diffusion coefficient. To explain these effects, we propose that in the presence of disorder, optical excitation produces electron-hole pairs which are localized in separate but closely correlated positions. If we assume that the electron and hole wave functions are localized on a scale length of order 100 Å and then also assume the lifetime is simply related to the wave function overlap, we can estimate that the separation distance is of order 200 Å. Furthermore, using the excitation intensity at the minimum value of the lower DT curve (saturation point of the DT spectrum), we can estimate the resulting *e-h* density to be of order 6×10^{11} *e-h*/cm^2/layer (using 0.5 W/cm^2, taking 50% of the energy distributed over the first 10 layers, and an effective excitation lifetime of 5 μs. This would correspond to an average distance between *e-h* pairs on the order of 140 Å, the same order of magnitude as the *e-h* separation estimated based on the *e-h* pair lifetime. This number could be interpreted as the average distance between *localization* sites. While it is difficult to relate this to interface morphology, it is comparable to that reported by chemical mapping.[4] The stepwise two photon transition discussed in the above 3LS model may correspond to excitations of two closely correlated *e-h* pairs. The unexpected low saturation intensities for the 3LS and 2LS are due to the long lifetime of these systems and the finite number of localization sites (recall that the saturation intensity is $\approx\hbar\omega N_T/\alpha_0\tau$ where α_0 is the absorption coefficient and N_T is the density of the localization sites).

Our model also provides an explanation for the discrepancy between the FWM and the DT spectra. As is well known, the FWM signal strength is proportional to the contrast ratio of the excitation grating produced by $\mathbf{E}_1^*\cdot\mathbf{E}_1'$. The contrast ratio is reduced if the input beams saturate the system. At the beam intensities used in the measurement ($I_1,I_1'\sim0.5$ W/cm^2, $I_2\sim0.1$ W/cm^2), the density of excitation (associated with spectral hole) is well below the saturation level, however the excitation in the increased absorption region is partially saturated, causing a grating flattening and resulting in a reduced FWM response.

In summary, we have shown that the low intensity nonlinear optical response in GaAs multiple QW is greatly affected by the presence of disorder. The measurements show that the existing theoretical work is inadequate to explain the low intensity nonlinear response in these systems. Finally, the long life time may need to be taken into account in potential device applications.

This work was supported by the Air Force Office of Scientific Research. The authors wish to acknowledge their appreciation to Professor P. K. Bhattacharya for supplying samples.

[1] See, for example, S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Adv. Phys. **38**, 89 (1989).
[2] J. Hegarty and M. D. Sturge, J. Opt. Soc. Am. B **2**, 1143 (1985).
[3] C. Weisbuch, R. Dingle, A. A. Gossard, and W. Wiegmann, Solid State Commun. **38**, 709 (1981).
[4] See, for example, A. Ourmazd, D. W. Taylor, J. Cunningham, and C. W. Tu, Phys. Rev. Lett. **62**, 933 (1989).
[5] C. A. Warwick, W. Y. Jan, A. Ourmazd, and T. D. Harris, Appl. Phys. Lett. **56**, 2666 (1990); D. Gammon, B. V. Shanabrook, and D. S. Katzer, Phys. Rev. Lett. **67**, 1547 (1991).
[6] T. Takagahara, J. Lumin. **44**, 347 (1989).
[7] H. Wang, M. Jiang, and D. G. Steel, Phys. Rev. Lett. **65**, 1255 (1990).
[8] D. G. Steel and J. T. Remillard, Phys. Rev. A **36**, 4330 (1987).
[9] H. Wang and D. G. Steel, Phys. Rev. A **43**, 3823 (1991).
[10] D. A. B. Miller, D. S. Chemla, D. J. Eilenberger, P. W. Smith, A. C. Gossard, and W. T. Tsang, Appl. Phys. Lett. **41**, 679 (1982); W. H. Knox, R. L. Fork, M. C. Downer, D. A. B. Miller, D. S. Chemla, C. V. Shank, A. C. Gossard, and W. Wiegmann, Phys. Rev. Lett. **54**, 1306 (1985); N. Peyghambarian, H. M. Gibbs, J. L. Jewell, A. Antonetti, A. Migus, D. Hulin, and A. Mysyrowicz, Phys. Rev. Lett. **53**, 2433 (1984). See also papers cited in Ref. 1.
[11] The scan in Fig. 1(b) covers a larger spectral region than the data in Ref. 7. However, additional structure is seen at energies away from the spectral hole in Fig. 1(b) compared to Ref. 7 because the value of δ was set to in these data to enable the detection of slower components. In Ref. 7, δ was set to 100 kHz to ensure that only the simple excitonic component was detected.
[12] H. Wang and D. G. Steel, Appl. Phys. A **53**, 514 (1991).
[13] In these measurements, the amplitude or phase shift on the lock-in amplifier is measured as a function of the modulation frequency and can then be easily related to the relaxation time.