Nonlinear Optical Response of the GaAs Exciton Polariton

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High-resolution differential optical transmission measurements of a thin layer of bulk GaAs show anomalous behavior of the lowest energy exciton at low excitation density and temperature. The spectrum shows induced absorption with a superlinear dependence on excitation density with no detectable change in resonance frequency or linewidth. At a temperature dependent excitation density, the anomaly undergoes a transition to a normal response due to excitation-induced dephasing. The results are fully consistent with the polariton picture generalized to include exciton-exciton interactions. [S0031-9007(97)04803-5]

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The coupling of resonant optical radiation below the band gap of direct gap semiconductors is dominated by excitonic resonances which exhibit features similar to atomic systems. However, unlike isolated atoms where the resonant coherent nonlinear optical response arises due to simple saturation when the Rabi flopping frequency approaches the spontaneous emission rate, the nonlinear response of excitons has been shown to be more complex. At moderate excitation levels, the response arises due to many-body effects such as phase space filling and Coulomb interactions including exchange and screening (see [1]). At lower excitation levels (typically between 10^{15} and 10^{16} excitons/cm^3 in bulk materials), recent work has shown that strong exciton-exciton scattering leading to line broadening, referred to as excitation-induced dephasing (EID), dominates the nonlinear response [2]. In this paper, we show that in high quality thin films of bulk GaAs, the nonlinear optical response becomes anomalous at reduced excitation levels ($<10^{15}$ excitons/cm^3): A temperature dependent transition occurs leading to induced absorption, a reversal of the sign of the nonlinear response, and apparent multiphoton behavior. We show that the entire range of experimental results is accounted for by the exciton-polariton picture which is generalized to include exciton-exciton interactions.

Measurements were made on several high-quality molecular-beam-epitaxy grown GaAs layers (surrounded by $Al_{0.3}Ga_{0.7}As$ layers between 200 and 500 nm) of various thicknesses (0.2, 0.3, and 0.5 μm). The samples were mounted on a sapphire disk and placed in a helium immersion cryostat following removal of the substrate by chemical etching to allow for optical transmission. The lh-hh (light-hole–heavy-hole) degeneracy was lifted by strain [3,4] as shown in the absorption spectrum in Fig. 1(a); the redshift is larger for the lh. The nonlinear response was determined using both cw frequency domain and transient nondegenerate differential transmission (DT) where the excitation and probing beams were independently tunable. The angle between the two beams in the laboratory was $\sim 5°$. The frequency domain DT measurements were made using two frequency-stabilized Coherent 699 Styryl 9 dye lasers (with linewidths of 5 neV) and time domain DT using a subpicosecond Ti:sapphire laser system. For the frequency domain measurements, the pump wavelength is fixed, for example, at the lh-exciton resonance, and the DT spectrum is obtained by scanning the

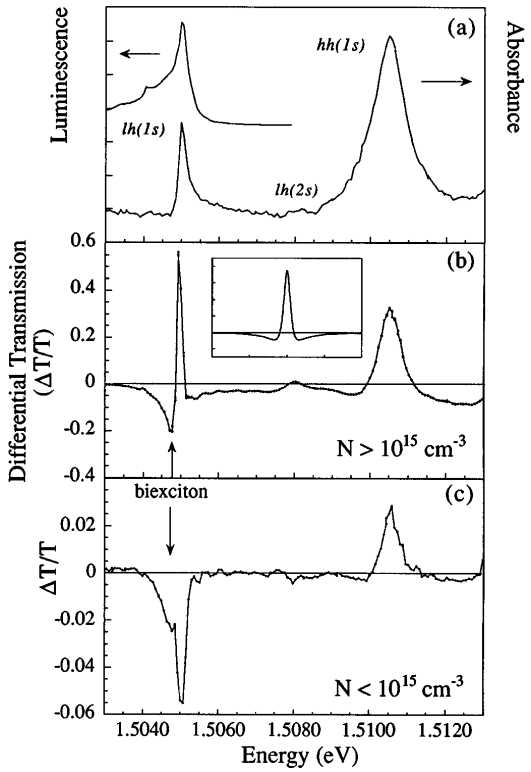

FIG. 1. (a) Absorption and luminescence spectrum of 0.2 μm bulk GaAs. Full width of lh($1s$) resonance is 0.2 meV. (b) DT spectrum at pump intensity of 116 W/cm^2 ($\approx 7 \times 10^{15}$ cm^{-3}) at 4 K, where the line shape is indicative of line broadening (and a redshifted induced absorption response attributed to the biexciton, indicated by arrow). Inset: theoretical line broadening DT curve, for comparison. (c) DT at a pump intensity of 13 W/cm^2 ($\approx 8 \times 10^{14}$ cm^{-3}) at 1.8 K, where induced absorption is clear at lh($1s$) state. Note line broadening line shape for the hh state. Probe intensity was 0.8 W/cm^2. Pump was resonant with lh($1s$).

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probe beam frequency. Both transmission and reflection of the probe beam were monitored using phase-sensitive detection following modulation of the pump beam.

The DT signal at $T = 4$ K at moderate exciton densities determined by the pump intensity ($>10^{15}$ cm^{-3}, estimated from the absorption, a 200 μm spot size and excitation lifetime) is shown in Fig. 1(b). The data is similar to that reported earlier [2], where the lh exciton clearly shows the DT line shape expected for EID. However, at lower excitation levels corresponding to exciton densities below approximately 10^{15} cm^{-3}, the DT response undergoes a transition and becomes completely anomalous as seen in Fig. 1(c): *An increase in absorption is observed across the entire lh-exciton absorption profile.* The peak response of the excitonic state remains unshifted in energy [a redshifted contribution seen in Figs. 1(b) and 1(c) arises from the biexciton [5]] and the linewidth remains approximately constant, independent of the excitation level in this regime. The reduction in transmission at the lh seen in Fig. 1(c) can be as much as 20%. The response of the hh($1s$) state is normal in the data shown, but also becomes anomalous for densities $<10^{14}$ cm^{-3}. Similar behavior was seen in several GaAs samples and additional measurements confirm that these signals are not artifacts.

Figure 2 shows the dependence of the DT signal at the peak of the lh($1s$) resonance at 4 K as a function of pump intensity (and hence excitation density), clearly illustrating the smooth transition behavior between induced absorption (giving a negative signal, indicating a reduction in transmission) and line broadening (giving an increase in transmission on resonance). Moreover, the induced absorption signal *increases superlinearly* with pump intensity at low

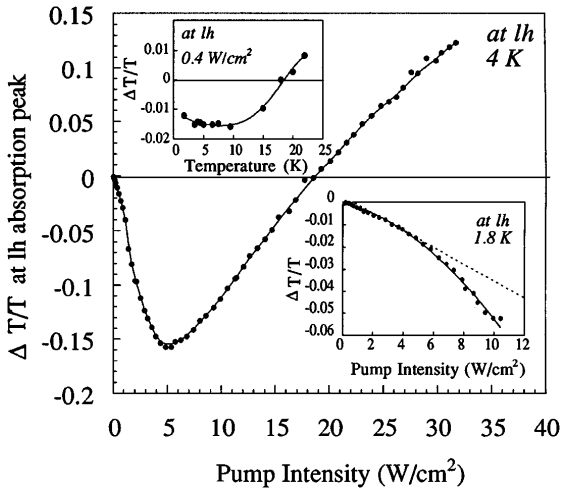

FIG. 2. Behavior of DT at peak of lh resonance at 4 K as a function of pump intensity. Probe intensity was 0.8 W/cm^2. Lower inset: superlinear behavior of lh($1s$) DT with increasing pump intensity; data taken at 1.8 K. Pump at lh($1s$). Dashed line given to clearly show deviation from linear dependence. Upper inset: dependence of induced absorption signal with temperature, with a pump intensity of 0.4 W/cm^2 and probe of 0.1 W/cm^2. Solid lines in curves are only a guide to the eye.

excitation levels when the pump is tuned to the lh($1s$) resonance, as shown more clearly in the lower inset of Fig. 2 at 1.8 K, where the superlinear dependence becomes more pronounced at lower temperatures. The initial curvature varies at least as I^2, suggesting a multiphoton interaction. This dependence becomes linear, however, when the pump is tuned, for example, into the continuum region.

The upper inset of Fig. 2 shows that the occurrence of the anomalous behavior is strongly temperature dependent, and is observable only at temperatures below about 15 K. At temperatures above 15 K, the DT line shapes show the normal EID structure similar to that in Fig. 1(b).

The decay time was determined by transient DT to be $\sim$0.5 ns (comparable to the excitation lifetime), with a pulse-width-limited rise time ($\sim$100 fs).

The present theoretical discussion of the nonlinear response of these systems has been highly successful in accounting for many experimental observations (see [1]). However, for the resonant DT response, the various contributions to the nonlinearity in the current models describe only a *decrease* in absorption at the peak of the excitonic resonance. We note that biexciton effects certainly lead to an increase in absorption, but this effect is redshifted as discussed earlier in this system [5] (see Fig. 1).

A physical and unified understanding of the behavior reported above can be obtained by a phenomenological generalization of the exciton-polariton model. It is well known that in the absence of strong exciton damping, the coupling between the exciton and the radiation field leads the formation of the exciton polariton [6–9]. In GaAs at low temperatures and excitation densities, polariton effects have been shown to be dominant, leading to distinct features in absorption [10], reflection [11,12], and luminescence spectra [12,13], as well as the direct observation of the polariton dispersion curve and group velocity [14,15]. The nonlinear optical effects of exciton polaritons in III-V quantum wells have also recently received attention [16].

In the polariton picture, a direct calculation of the absorption coefficient including the effects of dissipation and dispersion follows from the classical limit of a fully quantum calculation [17] with identical behavior seen in a fully classical picture [18]. As pointed out by Hopfield [6,19], the behavior of the polariton differs considerably from that expected for ordinary resonances in the absence of strong radiation coupling. In particular, for a simple isolated resonance (e.g., a harmonic oscillator or a two level atom), the absorption line shape is a Lorentzian with a width determined by the dissipation or dephasing rate. On resonance the absorption decreases with increasing dissipation rate while the linewidth increases. For an exciton polariton with weak dissipation, the absorption line shape is non-Lorentzian with a width determined by the strength of the exciton-radiation coupling. On resonance, increased dissipation leads to an increase in absorption with no change in the linewidth. As the dissipation rate increases further, the effect of the coupling between the exciton mode and the

radiation mode decreases and the absorption feature evolves into that of an isolated oscillator.

A formal description of this behavior is seen in the model of Akhmediev [18] where the absorption coefficient is determined by the complex index of refraction given by

$$n^2 = \varepsilon(\omega, k) = \varepsilon_b + \frac{f\omega_{\rm ex}^2}{\omega_{\rm ex}^2 - \omega^2 - i\omega\gamma + \beta c^2 k^2},$$

where $k = n\omega/c$, f is the exciton oscillator strength, γ is the dissipation rate, and $\beta = \hbar\omega_{\rm ex}/m_{\rm ex}^* c^2$ describes the exciton spatial dispersion. The critical damping parameter, above which the polariton effects disappear and the absorption formula takes the form of the familiar uncoupled oscillator result, is given by $\gamma_{\rm crit} = 2\sqrt{f\beta}\,\omega_{\rm ex}$. For GaAs, it is estimated that $\hbar\gamma_{\rm crit} \approx 0.3$ meV [11] [compared to the 0.2 meV full width in Fig. 1(a)]. We note that this value is greater than the longitudinal-transverse (LT) splitting in GaAs $E_{\rm LT} = (f/2\varepsilon_b)\hbar\omega_{\rm ex} = 0.08$ meV [11,15,17], which is sometimes taken as the measure for the importance of polariton effects (see discussion in [20]). For completeness, we note that a fully quantum discussion of absorption in the exciton-polariton picture including effects of dissipation is given by [21,22]. The results are similar to the classical models, however, exciton spatial dispersion is not included and the critical parameter that is developed in [21] is over an order of magnitude larger than that of the classical models and that reported in experimental studies [23].

The dramatic behavior described above as a function of dissipation has been reported in several semiconductors where the absorption features of the exciton show polaritonic structure in the absence of strong damping. With increasing dissipation due, for example, to phonon or free carrier scattering, the absorption on resonance was observed to increase while the linewidth and line shape remain nominally unchanged [23–26]. Above the critical damping rate, normal excitonic behavior was observed (i.e., simple line broadening with increasing dissipation and a decrease in absorption). Similar behavior is observed in our material, where, in the case of the lh exciton, increasing the temperature from 2 to 10 K leads to an increase in phonon scattering resulting in an increase in the lh-exciton absorption, with no change in spectral structure. The peak absorption becomes a maximum near 15 K and then decreases as the linewidth increases. Hence, the lh exciton in Fig. 1(a) is clearly polariton in nature.

To generalize the polariton absorption picture to describe the DT experiments above, we note that based on Fig. 1(b), the leading term in the nonlinear response under these conditions is excitation-induced dephasing, as expected from earlier experiments. Following the approach in [2,27,28] we take the linewidth γ to depend on the excitation density: $\gamma = \gamma_0 + \xi N$, where γ_0 is the dephasing rate at temperature T at zero exciton density, and ξN is the additional dephasing due to exciton-exciton interactions at exciton density N. In the polariton picture, we note that exciton-exciton scattering refers to the exciton component of the polariton [6,29–31]. Inserting this γ into the above expression for the complex index of refraction and solving for the absorption coefficient (recognizing that the density N is itself determined by the optical intensity and the absorption coefficient) we numerically determine the DT response as a function of laser intensity following the assumptions given in [18] for treatment of the two dispersion branches. The results are shown in Fig. 3 and qualitatively account for all of the data.

Specifically, Figs. 3(a) and 3(b) show the DT line profile as a function of decreasing pump intensity. The line shapes show a clear transition from line broadening due to EID in Fig. 3(a) to induced absorption at low excitation density in Fig. 3(b), where the dissipation rate is less than the critical rate, in excellent agreement with Fig. 1. Figure 3(c) shows the behavior of the peak DT signal as a function of pump intensity when the pump is tuned to the exciton-polariton resonance. The model reproduces the increase in absorption with a transition to line broadening giving a decrease in the peak absorption. The lower inset of Fig. 3(c) shows the initial superlinear dependence of the induced absorption signal on pump intensity when the pump *and* probe are tuned to the *same* resonance. Based

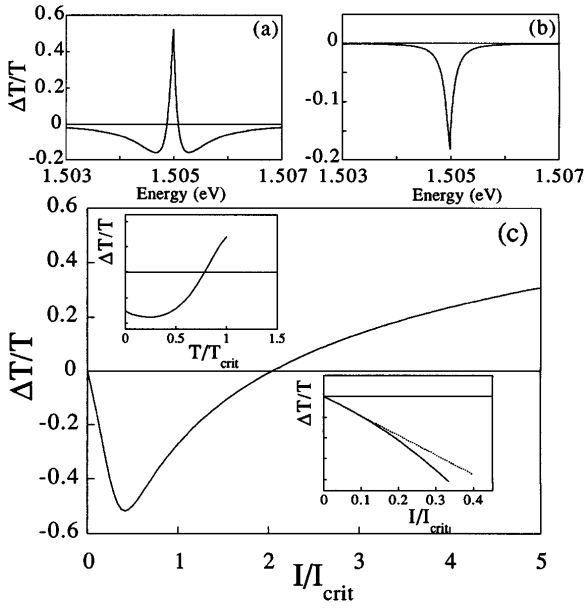

FIG. 3. Calculated DT based on the classical polariton model with spatial dispersion for increased damping due to increased exciton density. (Only changes in absorption are plotted since it was verified in the model that changes in reflection lead to only small corrections in the DT spectrum.) Spectra of DT for (a) $\gamma/\gamma_{\rm crit} = 4$, (b) $\gamma/\gamma_{\rm crit} = 0.4$. (c) Behavior of DT at absorption peak as a function of pump intensity. Lower inset: superlinear behavior of DT for pump and probe at *same* resonance. Upper inset: theoretical dependence on temperature T. In above, $I_{\rm crit}$ and $T_{\rm crit}$ are constants defined as the values at which $\gamma = \gamma_{\rm crit}$, calculated with the zero density and temperature absorption coefficient (with $I_{\rm crit} = 24$ W/cm^2 and $T_{\rm crit} = 14$ K). Values used in the calculation are $\hbar\gamma_0 = 0.1$ meV, $\xi = 0.75 \times 10^{-16}$ meV/cm^3, $\zeta = 14$ μeV/K, $\beta = 1.8 \times 10^{-5}$, $\hbar\gamma_{\rm crit} = 0.3$ meV and $\varepsilon_b = 12.5$.

on previously published parameters for f [32] and ξ [27], the model accounts for the transition to line broadening due to EID around a density of 10^{15} cm^{-3}, where the dephasing rate becomes equal to the calculated critical rate corresponding to a linewidth of approximately 0.3 meV for the lh.

The upper inset of Fig. 3(c) shows the theoretical dependence on temperature, where we have assumed a simple linear dependence for the intrinsic dissipation rate in agreement with independent measurements above 15 K. We take $\gamma_0(T) = \tilde{\gamma} + \zeta T$ corresponding to a simple model of single phonon scattering, where $\tilde{\gamma}$ is the value at $T = 0$. Based on a typical value for ζ [11,33] in agreement with the independently determined value for this sample, the model shows a transition temperature near 15 K, again in agreement with Fig. 2.

The above analysis of optical absorption in the polariton picture accounts for the increased absorption of the probe field at low pump intensity due to increased damping from exciton-exciton interactions with excitons excited by the resonant pump field. Since in this case the dissipation rate remains below the critical rate, there is no significant change in linewidth. The superlinear dependence on pump intensity in Fig. 2 is due to the fact that the pump absorption coefficient increases with increasing pump excitation. At sufficiently high excitation levels or temperatures, the damping rate due to exciton-exciton or exciton-phonon scattering exceeds the critical damping rate, overwhelming the exciton-photon coupling and polariton effects, leading to the ordinary excitonic nonlinearity of line broadening.

In summary, this paper describes the observation of an anomaly in the nonlinear optical response in GaAs which is clearly unexpected in the context of previous discussions of the excitonic nonlinear optical response. However, all of the essential features of the experiment are completely described by a model that allows for strong exciton-photon coupling leading to a polariton and excitation-induced dephasing. This work reveals an interesting characteristic of exciton nonlinearities with implications in any system where the coupling to the reservoir can be made weak with respect to the coupling to the photon, such as in quantum confined cavities (for example, see [34]).

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