Direct Measurement of Exciton-Exciton Interaction Energy

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(Received 10 April 2009; published 1 July 2009)

We report a direct measurement of the spectral function (real and imaginary self-energy) of excitons with a repulsive interaction potential. These results allow a stringent test of many-body theories of the exciton-exciton interaction which is independent of the exciton density calibration.

DOI: 10.1103/PhysRevLett.103.016403 PACS numbers: 71.35.−y, 78.67.De

The exciton-exciton interaction is notoriously difficult to determine both theoretically and experimentally. In contrast to atoms, for which the s-wave scattering length is known to high precision, after 50 years of study a definitive understanding of the exciton-exciton scattering cross section is still lacking. In the case of atoms, the nucleus can be treated as infinitely heavy compared to the electrons, and exchange between the nuclei can typically be ignored; for excitons, the positively charged particle (hole) has mass comparable to that of the electron, and one must take into account exchange not only between the electrons but also between the holes and also, in principle, between electrons and holes. The exciton is therefore in some ways analogous to positronium, but the ''gap'' energy of positronium is $2mc^2$, which is much larger than the interaction energies, while for excitons the gap energy is of the order of an eV, which is not so far different from other energies (typical Rydberg binding energies for excitons range from 0.01 to 0.1 eV). Numerous theoretical works [1–11] have estimated the exciton-exciton interaction for various assumptions and systems, but direct comparison of these predictions to experiment has been hard to come by.

The experimental situation has been nearly as difficult. The main difficulty is the calibration of the exciton density. Generating excitons involves absorption of photons with uncertainties in the absorption coefficient and the efficiency of exciton formation; detecting excitons via luminescence involves uncertainties in the rate of photon emission, the light collection efficiency, and the rate of nonradiative recombination processes. Intra-exciton-level absorption has received attention [12–14] recently as a method of calibrating exciton density, but it requires exciton levels which are well separated from each other and from other states, as well as calibration of the absolute absorption. In general, the overlap of the excitonic region of the spectrum with other electronic states such as electron-hole plasma, biexcitons, trions, and electron-hole liquid, each subject to both homogeneous and inhomogeneous broadening, can make it difficult to identify a purely excitonic signal.

In this Letter, we present experimental results for a measurement of the exciton-exciton interaction which are entirely independent of calibration of the exciton density. Our results can be compared to theories of the exciton-exciton interaction without needing to make any assumptions about the absolute exciton density. This is done by establishing a relation between the spectral width and the spectral shift, which is a function of temperature. Our results show that the present level of theory is not adequate to explain the data.

One thing which makes these experiments much simpler than many previous experiments is the design of the samples, which produces excitons with very long lifetime (compared to their thermalization time), completely repulsive interactions, and no other nearby states. This structure, known as a coupled quantum well, consists of two quantum wells, in our case GaAs quantum wells with 120 Å thickness, with a thin, 40-Å AlGaAs barrier between them. When an electric field is applied perpendicular to the plane of the wells, the electrons and holes move into adjacent wells and form spatially indirect excitons. Since the carriers need to tunnel through a barrier to recombine, they have an extremely long lifetime, in our case around 10 μs. This allows us to use a standard time-gated, intensified CCD camera with 5 ns time resolution to record the exciton luminescence long after the excitons have been created by a laser and have equilibrated to the lattice temperature. The spatial separation between the carriers also means that the interaction is completely repulsive [15]. This means that there are no other nearby spectral lines corresponding to other states such as biexcitons, trions, or electron-hole liquid. This simplifies the analysis tremendously, since in single quantum wells or bulk GaAs it is difficult to isolate the purely excitonic luminescence from that of other states at high density, while in our experiments there is a single line. The indirect exciton binding energy in these structures is approximately 4.5 meV [16].

The basic experiment is to create indirect excitons and allow them to come to equilibrium with the lattice and to measure the blueshift and broadening of the exciton spec-

trum over a wide range of densities. The shift and the broadening both depend on the same exciton-exciton interaction potential. Since both are linear with density, the ratio of the two will be independent of density. This ratio can be measured as a function of temperature and the result compared to theories of the exciton-exciton interaction.

Figure 1(a) shows typical spatially resolved spectra at low density and high density, both taken at times long after a quasi-cw laser pulse (chopped diode laser) with photon energy tuned to the direct (single quantum well) absorption resonance. The near-resonant excitation prevents any charge-separation effects which are observed when the excited carriers have high excess energy [17]. The excitons have been confined in an in-plane harmonic potential, as described in Ref. [18], to prevent diffusion out of the observation region. Figure 1(b) shows spectra extracted from the center of a similar trap, with 20 μm spatial resolution. These spectra were taken at different times, as the density decreased due to exciton recombination. Since the tunneling of carriers through the outer barriers of the wells was very slow compared to the repetition period of the laser pulses, the voltage across the wells and the leakage current through the sample were essentially constant in time and the same for all exciton densities. It is important to use this method for varying density, in which the time-averaged laser power remains constant, because varying the laser power can change the tunneling current, which in turn changes the effective voltage across the wells, which in turn can give a shift of the exciton line

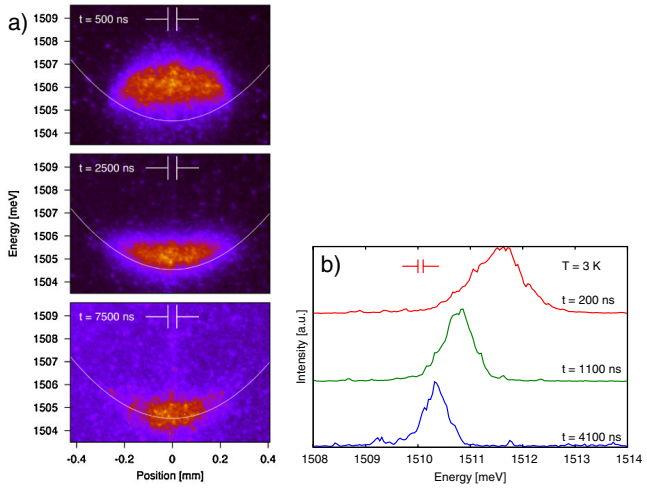

FIG. 1 (color online). (a) Spatially resolved luminescence spectra from indirect excitons in coupled quantum wells at $T = 5$ K, confined in a two-dimensional trap. (b) High-resolution spectra of the indirect excitons at the center of a similar trap, for three times after the laser excitation pulse, corresponding to different densities. There is an overall energy shift of the late-time spectral energy position relative to that of (a) because a slightly lower applied voltage was used. The integration window is 300 ns in all cases, much shorter than the exciton lifetime, 5000 ns in this case. All three spectra are normalized to unity.

unrelated to the exciton interactions. In these experiments, the leakage current was very low, about 0.1 μA/cm^2.

As seen in Fig. 1, there is a distinct spectral shift and broadening with increased density. The shift is entirely to the blue, as expected for repulsive exciton-exciton interactions. The spectral linewidth and maximum can be found by the best fit of a simple peak function to the data. In Fig. 2, we plot the blueshift as a function of the total spectrally integrated intensity at the center of the trap. The shifts are measured relative to the $t \rightarrow \infty$ value, that is, the zero-density limit reached at times long after the laser pulse. At long times after the laser pulse, the luminescence intensity can be taken as simply proportional to the exciton density, because the radiative decay rate of the excitons is constant. This is because the temperature of the exciton gas does not change in time once the excitons have reached equilibrium with the lattice [18].

In Fig. 2, the half width at half maximum of the spectral line is plotted as a function of the blueshift, for two temperatures. (Because the lifetime of the excitons depends on temperature, the applied electric field was adjusted to keep the lifetime the same at all temperatures. The electric field was still large enough in all cases, however, to keep the lowest exciton state strongly in the spatially indirect limit.) At late times, the linewidth becomes constant. This zero-density limit is determined by the inhomogeneous broadening due to disorder and charged impurities in the sample, as well as the homogeneous broadening due to exciton-phonon interaction [19,20], which depends on the temperature but not the exciton density. Our values of the low-density linewidth of the order of 0.5 meV are extremely low for a narrow quantum well sample and correspond to high carrier mobility [21]. As density increases, one theory [22] predicts that the line should narrow slightly due to filling of potential minima in the disorder landscape by the excitons, but we have not seen this effect under any conditions.

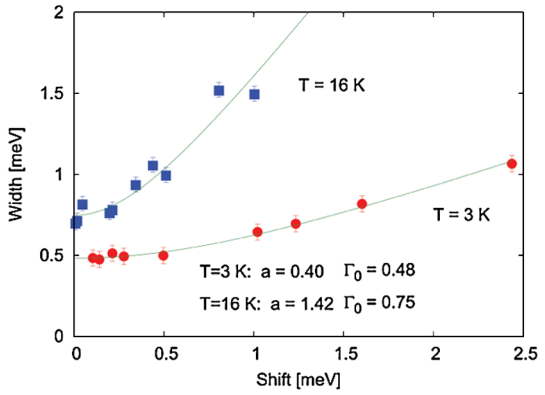

FIG. 2 (color online). Half width at half maximum of the exciton luminescence line as a function of the spectral shift at two temperatures. The solid lines are fits of the theory discussed in the text to the experimental values.

The data in Fig. 2 are fit to the formula $\Gamma = \sqrt{\Gamma_0^2(T) + [a(T)\Delta]^2}$, based on the assumption that the homogeneous broadening is convolved with the zero-density linewidth $\Gamma_0(T)$ and that the homogeneous broadening and the blueshift Δ are both linear with the density, and therefore the homogeneous broadening is proportional to the shift with some constant of proportionality $a(T)$. As seen in this figure, at high density the increase of linewidth is consistent with a linear dependence on the blueshift. Note that to get the unitless constant of proportionality $a(T)$, we do not need to make any assumptions about the absolute exciton density. In Fig. 3, we plot this constant as a function of temperature. This unitless function is a general result which should provides a test for many-body theory of the interactions.

A straightforward, second-order theory of the interaction can be written using an interaction of the form

$$H_{\text{int}} = \frac{1}{2A}\sum U(|\vec{k}_1 - \vec{k}_1'|)a^\dagger_{\vec{k}_1'}a^\dagger_{\vec{k}_2'}a_{\vec{k}_2}a_{\vec{k}_1}, \qquad (1)$$

where we use boson operators $a^\dagger_{\vec{k}}$ and $a_{\vec{k}}$ for the excitons [23]. Because the trap used in the experiments is macroscopic ($\sim$100 μm compared to the exciton Bohr radius of around 150 Å), a local density approximation can be used. The self-energy, to second order in U, is given by [24]

$$\begin{aligned}\langle E(\vec{k}_1)\rangle_T &= \frac{N}{A}\sum_{\vec{k}_2}[U(0) + U(|\vec{k}_1 - \vec{k}_2|)]\frac{e^{-E_2/k_BT}}{Z} \\ &\quad + \frac{N}{A^2}\sum_{\vec{k}_2,\vec{k}_2'}\frac{|U(|\vec{k}_1 - \vec{k}_1'|) + U(|\vec{k}_1 - \vec{k}_2'|)|^2}{E_1 + E_2 - E_1' - E_2' + i\eta} \\ &\quad \times \frac{e^{-E_2/k_BT}}{Z} \\ &\equiv \Delta_1 + \Delta_2 - i\Gamma, \qquad (2)\end{aligned}$$

where Z is the partition function. Using the Dirac theorem $1/(E - i\eta) = \mathcal{P}(1/E) + i\pi\delta(E)$, the imaginary second-order term becomes simply $\Gamma = \hbar/2\tau$, where $1/\tau$ is the out-scattering rate $(2\pi/\hbar)\sum|U(|\vec{k}_1 - \vec{k}_1'|) + U(|\vec{k}_1 - \vec{k}_2'|)|^2 n(E_2)\delta(E_1 + E_2 - E_1' - E_2')$, with $n(E) = ne^{-E/k_BT}/Z$ and $n = N/A$. The two real terms lead to an energy shift of the exciton luminescence line, while the imaginary term results in Lorentzian line broadening of the form $I(\Delta E) = 2\Gamma/[(\Delta E)^2 + \Gamma^2]$. This second-order approach has been used successfully before to fit the line shift and broadening in semiconductors due to interaction with phonons [25,26]. Reference [20] found the contribution of exciton-phonon interaction in GaAs quantum wells to the line broadening to be much less than 1 meV, consistent with our zero-density data. The line shapes in our experiments are not perfectly Lorentzian and have a slight asymmetry at high density, as seen in Fig. 1(b). When quantum statistics at high density come into play, this second-order approximation breaks down, as discussed by Zimmermann [27]. In the experiments reported here, the exciton density (for which we have an experimental upper bound based on the total photon absorption) was kept well below the density range where quantum statistics should come into play.

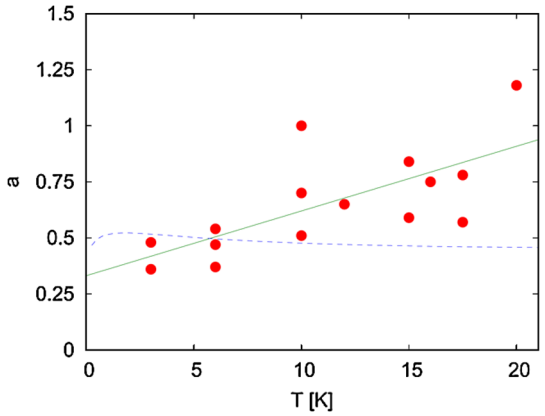

FIG. 3 (color online). The unitless slope of the spectral broadening vs blueshift, from data fits like those of Fig. 2, as a function of temperature. The green line is the best fit to a straight line, which yields $a = (0.029 \pm 0.008)T/K + (0.33 \pm 0.10)$. The dashed blue line is the prediction of Ref. [11].

A natural assumption is to take U as constant over the range of k's occupied by the excitons, since the average thermal k at low temperature is smaller than π/a_B, where a_B is the exciton Bohr radius. In this case the second-order real term vanishes, and the shift and broadening become simply

$$\Delta_1 = 2Un, \qquad \Gamma = 4\pi|U|^2 n\mathcal{D}(\bar{E}), \qquad (3)$$

where $\mathcal{D}(\bar{E})$ is the density of states per unit area at the average thermal energy. In these two-dimensional structures, the density of states is constant and equal to $gm/2\pi\hbar^2$, where g is the spin degeneracy.

This straightforward approach, which works well for exciton-phonon scattering, gives results which are strongly out of accord with our exciton-exciton scattering data. If the mean-field, first-order term Δ_1 is taken as responsible for the observed blueshift of the data of the order of 1 meV at densities around 10^{10} cm^{-2}, the implied value of the constant U gives a broadening of greater than 10 meV, much more than the observed values. It also implies that both Δ and Γ are independent of T, in disagreement with the dependence seen in Fig. 3.

An improvement can be made by using a T-matrix approach to take into account many-body correlations of the excitons [11]. This approach gives the broadening and blueshift of the luminescence comparable to each other for realistic calculations of the coupled quantum well exciton interaction, in agreement with experiment. However, this theory also predicts that the ratio of the broadening to the blueshift should be a decreasing function of temperature,

while our experiments show that the ratio of the broadening to the blueshift increases with temperature. Part of the reason for the discrepancy may be a numerical error in taking the ratios of two calculations. Both the blueshift and the broadening (real and imaginary self-energy) in Ref. [11] increase with temperature, but, according to those calculations, the blueshift increases faster with temperature than the broadening. A change of the numerics at low temperature might give a different ratio between the two.

Another recent theory [28], which uses a similar approach to Ref. [11], implies that the blueshift should be proportional to $nT^{1/3}$ in the low-density limit and that broadening due to fluctuations should be proportional to $n^{1/2}T^{2/3}$. This gives the temperature dependence of the ratio of the broadening to the shift proportional to $T^{1/3}$, which is consistent with the increase seen in Fig. 3, but the $n^{-1/2}$ density dependence of the ratio is not consistent with our observations.

In conclusion, these experiments, made possible by the extremely low inhomogeneous broadening and disorder in these samples as well as the extremely long exciton lifetime due to the coupled quantum well design, give a clean set of data to compare to theories of the exciton-exciton interaction, independent of any ambiguities of exciton density calibration. Understanding the exciton-exciton interaction is crucial for many nonlinear optical effects and many-body theories of exciton behavior. Although basic second-order theory of the self-energy of the excitons implies the effects of blueshift and broadening for excitons with repulsive interactions, a quantitative calculation of these effects requires a fairly sophisticated many-body theory. However, such a theory is not a multiparameter theory, because we have a single, isolated spectral line which has shape and position, i.e., spectral function, which depend entirely on the interaction potential $U(k)$, which in turn depends only on known values of the electron and hole mass. The recent T-matrix theory [11] correctly gives the real and imaginary self-energy as comparable but gives an incorrect prediction for the temperature dependence.

This work has been supported by the Department of Energy under Grant No. DE-FG02-99ER45780. We thank R. Zimmermann, B. Laikhtman, and P. Littlewood for helpful discussions.

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