

Huge density-dependent blueshift of indirect excitons in biased coupled quantum wells

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We measure a dynamical blueshift of the indirect exciton in GaAs-coupled quantum wells of over 20 meV for carrier densities in the range 10^9 – 10^{12} cm $^{-2}$. We show that this shift is a many-body effect, which is proportional to the indirect exciton density over a broad range of densities, and it is accompanied by homogeneous broadening, which is proportional to the indirect exciton density.

A nonlinear Stark shift, in which the wavelength of an optical resonance depends on the intensity of an input optical beam, is of great interest because it can be used in various schemes of optical switching; if an input optical signal strongly changes the absorption of a medium then a second optical signal can be switched on and off in the optical analog of a transistor. This behavior, based on the ac Stark effect, has been demonstrated in GaAs single quantum wells^{1,2} and other systems^{3–5} and has been utilized in switching devices.^{6,7} A limitation of many versions of the ac Stark effect is that a strong light signal must be used to generate a relatively small shift. By analogy with the electrical transistor, what one would like is a device in which a weak input signal produces a large change in the output signal.

We report here a system in which the optical resonance wavelength depends extremely sensitively on the intensity of an input light beam. This occurs in conjunction with the dc Stark effect, also known as the quantum confined Stark effect, in coupled GaAs quantum wells. The effect is a manifestation of the many-body renormalization of the exciton energy levels due to their interaction, an effect that has attracted much theoretical interest over the years (e.g., Refs. 8–11.)

I. THE SPECTRAL SHIFT WITH DENSITY

Our samples consist of a single pair of 60-Å GaAs quantum wells separated by a 42-Å Al_{0.3}Ga_{0.7}As barrier, surrounded by thick, pure AlAs barriers. The substrate is heavily *p* doped and the capping layer is heavily *n* doped in order to allow electric field perpendicular to plane of the quantum wells. An electric field causes electrons to tend to become confined in one well and holes to tend to become confined in the other well; “indirect” excitons are formed from a conduction electron in one well and a heavy hole in the other well, while “direct” excitons are formed from an electron and heavy hole in the same well. As reported elsewhere by several authors, indirect excitons in coupled quantum wells have a strong dc Stark shift^{12–14} and long lifetime.^{15,16} In our samples the indirect exciton line shifts about 70 meV (40 nm) when the dc voltage across the sample is varied from 0 to 5 V (i.e., fields up to 60 kV/cm),

and the indirect excitons have lifetimes of about 100 ns at high applied field.¹⁷ The direct exciton luminescence line does not shift strongly with electric field; in these samples it lies at 765 nm.

For these experiments, we immersed the sample in liquid helium at 2 K and excited it with a laser tuned to the direct exciton resonance, focused to a 50 μm spot on the sample surface, and observed the indirect exciton luminescence with time resolution of 40 ps and spatial resolution of 10 μm. The laser pulse width was about 5 ps, with a spectral width on the order of 0.1 meV, and the repetition rate was 3.8 MHz. Figures 1 and 2 show the spectral shift of the indirect exciton luminescence line as a function of laser excitation intensity. The excitons in this case were trapped in a potential minimum produced by inhomogeneous applied stress as described previously,^{17,18} but the stress produces only an overall dc shift of the indirect exciton line and does not affect the nonlinear blueshift. In Fig. 2(a), the indirect exciton line appears to shift as $\log(\text{density})$, but this can be understood more easily as a linear shift at low density of 5×10^{-11} meV cm 2 which saturates to a linear shift of 1.5×10^{-13} meV cm 2 at high density, as seen in Fig. 2(b). The low-density shift is almost 20 times larger than the shift reported at an estimated density of 10^{11} cm $^{-2}$ for excitons in single quantum wells;¹⁹ as seen in Fig. 1, the total shift of 25 meV is much larger than the typical linewidth of about 3 meV. The laser powers used here are extremely low. Measureable spectral shifts were observed for average laser powers as low as one microwatt.

We do not have an independent measure of the electron-hole pair density, but the laser photon intensity provides an upper bound. Since the substrate luminescence in the range 800–810 nm is much stronger than the indirect exciton luminescence, we know that most of the laser light is not absorbed by quantum wells. We estimate that the maximum pair density for a given laser intensity is a few percent of the value for the excitation density given on the bottom axis of Fig. 2. This is because if the density were higher, Fermi state filling of the conduction electrons would lead to significant broadening of the luminescence line, while as seen in Fig. 1, the luminescence has broadening less than 3 meV, except at very early times after the laser pulse, as discussed below. When the excitation density exceeds 10^{14} cm $^{-2}$, free

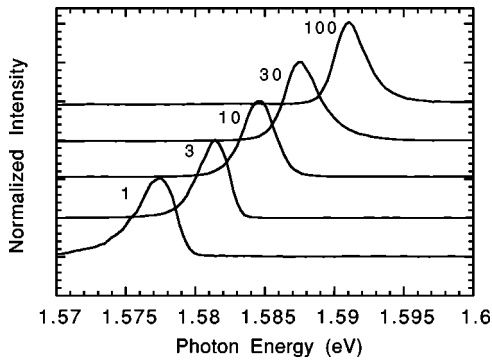


FIG. 1. Time-averaged luminescence spectra for various laser intensities, for a 60-42-60 Å GaAs/Al_{0.3}Ga_{0.7} coupled quantum well structure immersed in liquid helium and excited by a laser tuned to the direct exciton resonance as described in the text. The applied electric field was 61 kV/cm; in addition, applied stress shifted the line 5 meV lower as discussed in Ref. 17. The labels of the curves are the average laser intensity in W/cm².

electron-hole recombination luminescence is seen as a separate line in the luminescence, indicating that at lower densities, the luminescence comes almost entirely from the indirect excitons.

There are several possible mechanisms for a nonlinear shift of the exciton ground state. The ac Stark effect studied by several authors in the past (e.g., Refs. 1 and 2, cf. Ref. 20) is essentially a renormalization of the excitonic states due to the intense electric field of a laser tuned near the excitonic resonance. As such, it ends when the laser field is gone. In the present experiments, the Stark shift of the excitonic states persists long after the laser pulse, with a lifetime on the order of the exciton lifetime. There are two ways to envision a density-dependent shift in this case. One possibility is that free electrons and holes created by the laser screen out the perpendicular electric field or screen out the electron-hole Coulomb interaction. A second possibility is that the energy shift is primarily a many-body effect of the exciton-exciton interactions. Previous work^{17,19} indicated that free carriers played an important role; in particular, in Ref. 17 it was shown that at high excitation intensity the direct excitons have shorter lifetime immediately after the laser pulse, which would imply that the electron-hole wave-function overlap had increased, i.e., the electric field felt by the excitons was reduced by screening. Free carriers are a natural candidate for causing this screening. In both of those studies,^{17,19} the laser created carriers at energies well above the excitonic ground state, and therefore a substantial population of free carriers could exist. In the present case, the laser is tuned directly to the direct exciton state, so that no free carriers are created. This is confirmed by the fact that we see only two luminescence lines from the quantum wells at all times, corresponding to the direct and indirect excitons, with no extra lines for free carrier recombination, which should appear about 10 meV higher than the direct exciton line for intrawell recombination²¹ and about five meV higher than the indirect exciton line.^{13,22} At all but the very earliest times after the most intense laser pulses, the homogeneous broadening of the exciton lines is low enough that a free carrier recombination line should be easily resolvable. If there are no free carriers, then the screening and state renormalization

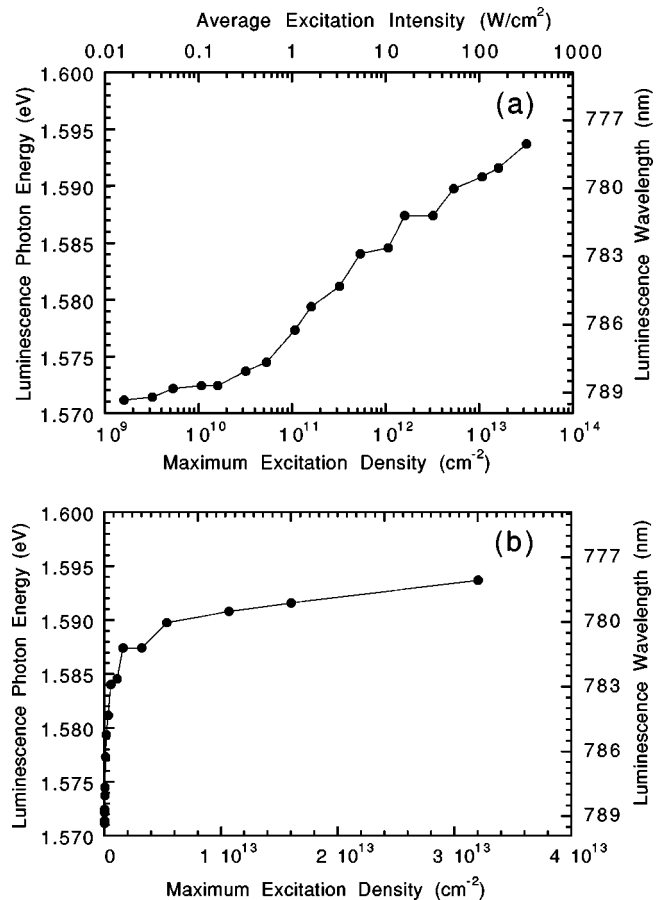


FIG. 2. (a) The energy of the indirect exciton luminescence maximum as a function of laser intensity, for same data shown in Fig. 1. (b) The same data plotted on a linear density scale. The shift of the line at low density is 5 meV/W cm⁻², or 5×10^{-11} meV cm² in terms of the peak excitation density.

must come from the excitons themselves.

If the shift comes from the interactions of the excitons with each other, there are several different ways to envision how this comes about. One picture is that the screening of the excitons by each other reduces the binding energy of the excitons.²³ In the present experiments, the shift of the line is much larger than the exciton binding energy, so this cannot be the primary mechanism for the blueshift. Another picture is that the excitons screen out the perpendicular electric field, which reduces the quantum confined Stark shift.¹⁹ Another picture is that the long-range van der Waals attraction of the excitons is screened out, leaving a greater overall repulsion between the excitons due to Pauli exclusion.²⁴ Last, one can view the excitons as repulsive point particles²⁵ and view the blueshift as due to the mean-field interaction self-energy. In our opinion, these effects can not be considered separately, and a proper theoretical treatment would start with the full electron-hole Hamiltonian and include all Coulomb interactions.

We have not yet studied the dependence of the effect on the excitation wavelength in detail, but it is clear from our experiments so far that the blueshift is much weaker when the excitation is tuned to wavelengths in the 600 nm range, with photon energy well above the direct exciton resonance. This would seem to imply that the creation of free carriers

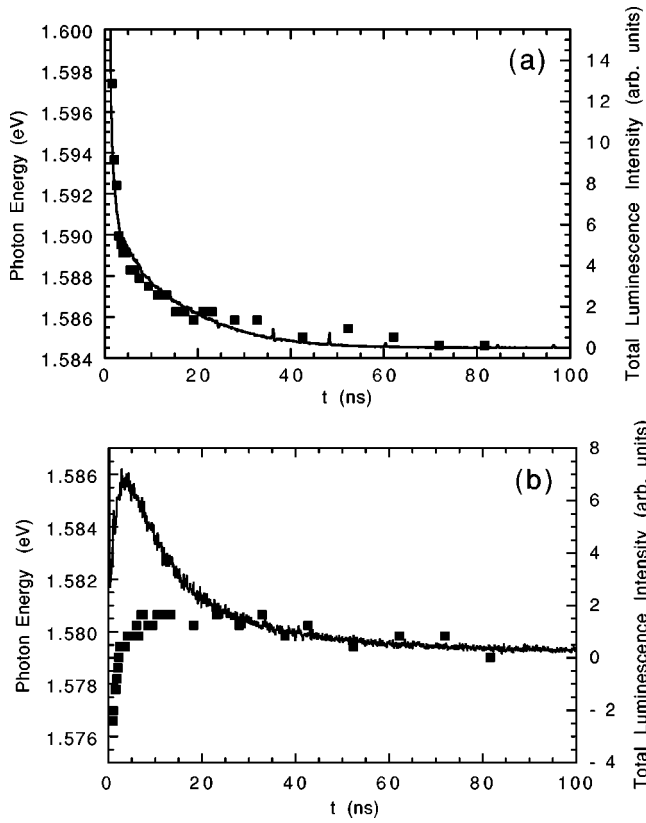


FIG. 3. (a) Squares: the energy of the indirect exciton luminescence maximum as a function of time following a short (5 ps), intense (1.5 nJ) laser pulse tuned to the direct exciton resonance (left-hand vertical axis), for the same structure and experimental conditions as in Fig. 1. Solid line: the total luminescence intensity at the same times (right-hand axis). (b) The same as (a) but for a much weaker (0.023 nJ) laser pulse.

actually *reduces* the Stark shift instead of enhancing it. This can be understood if one assumes that the free carriers primarily act to screen out the interexciton interaction, which as discussed above is the primary cause of the blueshift and the homogeneous broadening. It may also be that the higher photon energy leads to greater lattice heating, with the consequent redshift of the band gap, as discussed below.

II. TIME-RESOLVED RESULTS

If the blueshift is proportional to the exciton density as expected for a mean-field effect, then one would expect to see this in the time-resolved spectra as the density of the excitons decays. Figure 3(a) shows the energy of the indirect exciton luminescence as a function of delay after an intense laser pulse (squares, left-hand axis). Plotted on the same figure is the total luminescence intensity (solid curve, right-hand axis), which is proportional to the indirect exciton density since the indirect exciton luminescence lifetime is constant after the first 2 ns. The blueshift of the indirect exciton line is clearly proportional to the indirect exciton density over a wide range of density.

The dynamics are more complicated than this figure might indicate, however. As seen in Fig. 3(b), for a much less intense laser pulse, the spectrum can actually shift both to the red and to the blue at different times. (The late-time energy

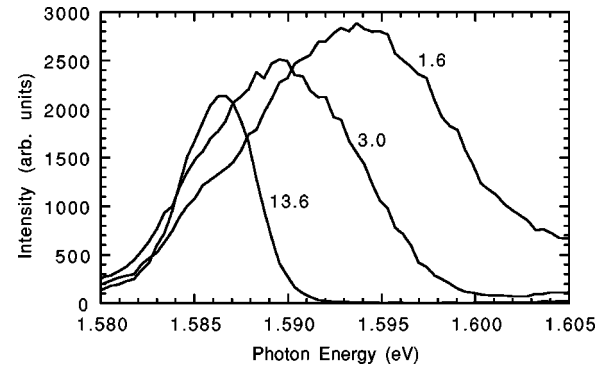


FIG. 4. The indirect exciton luminescence spectrum at various delays after an intense (1.5 nJ) laser pulse; the same data as in Fig. 3(a). The labels of the curves are the delay in nanoseconds after the laser pulse.

positions of these two cases do not coincide because there is a small cw leakage component from our laser, which has intensity proportional to the main pulse intensity.) There are two possible mechanisms for the observed redshift at moderate density. First, in addition to the density-dependent renormalization of the exciton states, there is a well-known redshift of the luminescence due to reduction of the GaAs band gap with increasing temperature.²⁶ The initial redshift of the luminescence in Fig. 3(b) can be interpreted as arising from the gap shift of the lattice, which relaxes quickly back to the bath temperature of 2 K. In the case of Fig. 3(a), this interpretation would imply that the initial blueshift should be much larger, but is partially canceled by the redshift due to lattice heating, consistent with the saturation of the blueshift discussed above. It is not surprising that the gap redshift and the mean-field blueshift depend differently on the laser power, because the band-gap redshift is approximately proportional to T^2 ,^{26,27} and the lattice temperature depends on the total heat energy U approximately as $U = CT^4$, i.e., $\Delta E \propto U^{1/2}$. It is not clear how quickly the lattice temperature relaxes back to the bath temperature in each case. In general, the rate at which the local temperature of the excited region relaxes is slower when more total heat is deposited, due to the “phonon hot spot”^{28–30} of elevated temperature, which can persist in a semiconductor up to hundreds of nanoseconds after an intense laser pulse.

A second mechanism for the redshift and blueshift at different times may be the temperature dependence of the density-dependent exciton state renormalization itself. As discussed in Ref. 8, the renormalization of the exciton states can lead to a blueshift or a redshift with density depending on their temperature. This is in addition to the renormalization of the single-particle band gap.

The interpretation of the blueshift as a mean-field effect is strengthened by examining the homogeneous broadening of the luminescence line. Figure 4 shows that for a high-intensity pulse there is also a large homogeneous broadening of the indirect luminescence, which decreases in time following the laser pulse. The full width at half maximum of the indirect exciton luminescence is plotted in Fig. 5 as a function of the total luminescence intensity. When the contribution of inhomogeneous broadening is removed, it is clearly seen that the homogeneous broadening is linearly dependent on the total indirect exciton density.

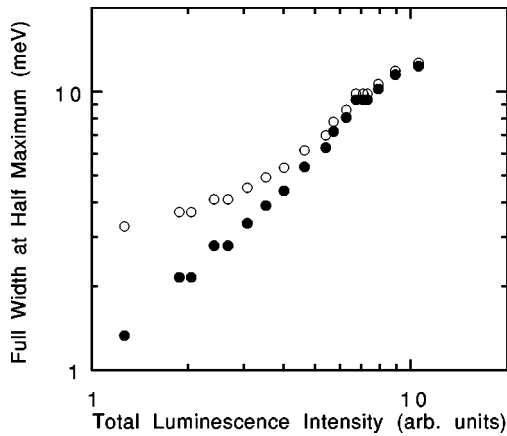


FIG. 5. Open circles: the full width at half maximum (FWHM) of the indirect exciton luminescence spectra shown in Fig. 4 as a function of the total indirect luminescence intensity at the same times. Solid circles: the homogeneous broadening deduced from the same data by taking $\Delta_{homog} = \sqrt{\Delta^2 - \Delta_{inhomog}^2}$, where Δ is the FWHM of the luminescence line and $\Delta_{inhomog}$ is the inhomogeneous broadening, taken as the late-time value of Δ , 3.0 meV in this case.

As seen in Fig. 1, at low densities, the blueshift is large, while the homogeneous broadening is much less than the inhomogeneous broadening. This is consistent with the results of Ref. 19, which found a measurable blueshift but negligible density-dependent homogeneous broadening. This is not surprising because the mean-field shift is linear in the interaction constant, while the homogeneous broadening, i.e., the imaginary self-energy, depends on the square of the interaction constant.³¹

Our interpretation of these data is therefore that at low densities, the blueshift of the indirect exciton line is a many-body effect caused by the large dipole-dipole interaction of the indirect excitons, with negligible redshift due to lattice heating, while at higher density, the average luminescence energy shifts much less strongly with exciton density at early times because the effect of lattice heating partially cancels the many-body blueshift. In the case of Fig. 3(a), the blueshift is proportional to the exciton density because the lattice temperature remains roughly constant over the first 50 nanoseconds, while in the case of Fig. 3(b), the lattice cools much more quickly.

Because the center of the laser spot is more intense than the outside, the density-dependent Stark shift of the indirect excitons also leads to spatial gradient in energy, i.e., a force. Figure 6 shows an image taken with a time-gated charge coupled device (CCD) camera on an imaging spectrometer, which shows the spectrum of the indirect excitons in the first 50 ns after the laser pulse as a function of position for a relatively defocused laser spot. The spectrum is broader in the center because of the density-dependent homogeneous broadening, and shifted to higher energy.

III. CONCLUSIONS

This effect has great promise for optical switching applications, because the shift appears to be directly related to the exciton-exciton interactions, which are intrinsically fast, instead of macroscopic, slow processes.

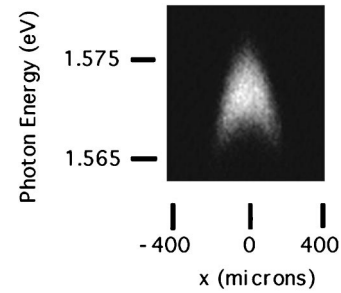


FIG. 6. Time-gated image of the indirect exciton luminescence for the first 50 ns after a short (5 ps) laser pulse, using a time-gated Princeton CCD camera on the back of an imaging spectrometer.

There are three present limitations to using this effect in optical switching. First, the fact that the electron and hole wave-function overlap is so low not only implies that the indirect exciton states have long lifetime; it also implies that the absorption of light by the indirect exciton resonance is very low. In order to increase the efficiency of switching schemes, multiple sets of coupled quantum wells can be used, or the system can be placed in a Fabry-Perot cavity, as in, e.g., Ref. 32.

Second, because the spectral shift depends on the exciton density, it persists to very late times, while many schemes of optical switching require fast extinction. This is because we have used above-gap excitation in these experiments. It is therefore important in the future to perform a standard optical Stark shift experiment on this type of structure in which the pump laser is tuned to a photon energy below the indirect exciton ground state, so that no real excitons are created. The fact that the shift of the indirect exciton line comes from many-body interactions of the excitons themselves, as shown here, is very significant, because this same interaction goes into the theory of virtual excitons,²⁰ which describes the optical Stark effect in the case of below-gap excitation. Because the exciton-exciton interactions are so huge in this system, a standard optical Stark shift experiment with below-gap excitation is also likely to show large shifts.

Finally, immersion in liquid helium is a severe limitation for technological applications. In this regard, we note that the effect reported here is a general one and does not depend on particular features of GaAs. The polarization of the excitons leads to a much stronger repulsion between excitons. Therefore, if a coupled quantum well structure is created, which allows the existence of excitons at room temperature, then this effect should occur at room temperature as well. It is well known that excitons in the semiconductor Cu_2O exist at room temperature³³ with a binding energy of 150 meV and an exciton-phonon dephasing time of approximately 150 fs, and therefore it would be valuable to look for a quantum confined Stark effect of excitons in Cu_2O quantum wells.

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