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Optical nonlinearities in multiple quantum wells: New insight on band-gap renormalization

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The contribution of many-body effects in the optical nonlinearity of excitons in multiple quantum wells is studied through optical pump-and-probe experiments. It is found that, when a dense electron-hole population is photoexcited, the renormalization of unoccupied subbands is relatively weak, in contrast to the strong effect observed in luminescence. This result points out the limitations of the rigid-shift model for band-gap renormalization.

The excitonic resonances of multiple quantum wells (MQW's) display very strong optical nonlinearities. The picture that has emerged in recent theoretical work[1-3] is that, to a first approximation, two types of optical nonlinearities can be distinguished according to their origin: those involving single-particle states (phase-space filling) and those that may be attributed to many-body interactions among the photoexcited species. The former arise because Pauli's exclusion principle applies to the electrons and holes constituting the excitons, and this produces a finite exclusion volume for each exciton. As electrons and holes fill their phase space, each additional exciton in the crystal has a smaller volume available: no more excitons can be created when their total (cumulative) exclusion volume becomes of the order of the crystal volume, and the excitonic resonance saturates. On the other hand, many-body interactions refer to the direct and exchange Coulomb interactions of electrons and holes, which at high plasma density give rise to effects such as screening and band-gap renormalization (BGR). These effects may cause broadening or even disappearance of the excitonic resonance (due to the dissociation of the excitons) as well as a spectral shift.

Usually, nonlinear-optical experiments show a cumulative effect due to all sources of nonlinearity simultaneously. Recent experimental work,[4] however, has helped sort out the contributions of phase-space filling from those of many-body effects: By optically injecting a short pulse (~ 150 fs) of electron-hole pairs distributed over a narrow energy range and by monitoring the temporal evolution of the spectrum of the MQW's as the carriers thermalized, Knox *et al.*[4] were able to establish that the dominant source of optical nonlinearity near the excitonic resonances is phase-space filling: it produces at least 6 times more bleaching than the many-body effects, which they termed collectively "screening." In the experiments reported in this paper we examine more closely the many-body effects occurring in photoexcited MQW; by optically injecting excitons in one subband (through a relatively long light pulse of 10 ps) and examining the spectral modifications of an unoccupied subband in the same time scale. These experiments give a direct experimental handle on the study of the BGR of the second electron–heavy-hole transition through due to a dense population of excitons in the first subband, and they permit us to address the problem of renormalization of unoccupied electronic states, far above the Fermi level. Several recent optical experiments have also examined many-body effects in low-dimensional structures.[5-7]

The light source consists of two optical parametric generators (OPG) pumped simultaneously by a frequency-doubled neodymium-doped yttrium aluminum garnet (Nd:YAG) laser that produces single pulses at 20-Hz repetition rate. The two OPG's can be tuned independently between 600 and 1700 meV (0.7–2 μm), and produce pulses of 10 ps duration, approximately 1 meV spectral width, and 100 MW/cm^2 peak intensity (1 mJ/cm^2 per pulse). The light source is described in more detail in Ref. 8.

The sample used in these experiments is a 60-period MQW grown by molecular-beam epitaxy (MBE) on a GaAs substrate, with 114-Å-wide GaAs wells and 79-Å-wide $Ga_{0.7}Al_{0.3}As$ barriers (measured by x-ray diffraction). The MQW region is sandwiched between two 1-μm-thick $Ga_{0.7}Al_{0.3}As$ layers. It has p-type residual doping of 10^{15} cm^{-3} (10^9 cm^{-2} in a layer). A 2-mm^2 hole was chemically etched in the GaAs substrate to allow optical transmission experiments in the MQW. The hole was located at the cleaved edge of the substrate, and thus the optically accessible part of the MQW was surrounded only on three sides by the substrate. The configuration was adopted so that the edge of the sample could be accessible to the pump beam, and thus minimize the reabsorption of the luminescence emitted at right angles with respect to the pump beam. Antireflection coatings of Al_2O_3 were deposited to eliminate Fabry-Perot effects. The experiments were performed at a temperature of 15 K by means of a closed-cycle He refrigerator.

Assuming that at the Γ point the conduction-band discontinuity is 60% of the band-gap difference between GaAs and $Ga_{1-x}Al_xAs$,[9] the band structure of this MQW near the fundamental gap consists of three electron subbands (E_n), four heavy-hole subbands (H_n), and three light-hole subbands (L_n). The binding energies of the excitons associated to the three lowest-energy transitions are 7.5 meV for the E_1H_1, 8.5 meV for the E_1L_1, and 6.5 meV for the E_2H_2 excitons.[10]

In the first experiment, the two beams produced by the two OPG's were both focused inside the sample. One of them, the pump beam, was tuned to the heavy-hole–exciton resonance of the first subband (E_1H_1), at 1540 meV. It was focused to approximately 250-μm diameter, thus attaining an energy density of 80 μJ/cm^2 per pulse, corresponding to an estimated photoexcited pair density of 3.5×10^{11} cm^{-2}. The other, the probed beam, was attenuated so as not to produce any detectable spectral saturation (i.e., at most 10^{10} cm^{-2} photoinduced carriers in a layer) and was focused to a diameter of 200 μm so that it probed only the central part of the pump. Its wavelength was scanned in order to measure the absorption spectrum of the sample. A portion of the probe beam was used as a reference, to correct the spectra for pulse-to-pulse intensity fluctuations by taking the ratio of the reference and the probe beams. The spectra obtained are displayed in Fig. 1.

The linear absorption spectrum of the sample (solid line) was measured by the probe beam in the absence of the pump beam. It shows the E_1H_1 and the E_1L_1 excitons at 1540 and 1547 meV, respectively, and the E_2H_2 exciton at 1620 meV. An additional peak, observed right next to the E_2H_2 exciton (1626 meV), is probably due to the mixing of the L_1 and H_2 bands.[11] The narrow well-defined excitonic lines show the high quality of our sample.

The nonlinear spectrum (dashed line) was obtained with the probe and the pump beams spatially coincident inside the sample, but with the probe pulse arriving in the sample just after the pump pulse ends (i.e., a peak-to-peak delay of 10 ps between the two pulses), to maximize the amount of pump energy deposited in the sample. When compared with the linear spectrum, we note that the excitonic resonances in this spectrum are modified. In particular, the E_2H_2 resonance shifts and broadens, while E_1H_1 and E_1L_1 vanish. In interpreting this nonlinear spectrum, we note that, due to the resonant pumping of the E_1H_1 excitons, the phase space of these states becomes filled. This produces strong optical saturation of the E_1H_1 and E_1L_1 resonances and masks the manifestations of the others sources of optical nonlinearity in this spectral region.[4] At the same time, the localization of bleaching in the spectral vicinity of the E_1H_1 and the E_1L_1 excitons indicates that the carriers are thermalized on the time scale of our experiment, and therefore all the states of the E_2 and H_2 subbands are free. This means that the E_2H_2 exciton is not subject to phase-space–filling effects, and that all the changes observed in its spectrum (i.e., a spectral broadening, a slight decrease of the area under the peak, and a red shift) must be due *only* to the many-body effects produced by the population of the first subband. In particular, the broadening of E_2H_2 (of about twice the unpumped spectral width) may be attributed to screening which modifies the structure of the exciton and/or to collisions with the dense population of the first subband which may cause dephasing or ionization of the E_2H_2 exciton. The small decrease of the area of the E_2H_2 exciton peak (between 10% and 30% depending on the model used for disentangling the spectral contributions of the excitonic lines and band-to-band transitions) indicates that the excitonic oscillator strength is not affected very much by the presence of a dense electron-hole population in the first subband, and attests to the relative inefficiency of quasi-two-dimensional (2D) screening.[12] Assuming that screening by the photoexcited electron-hole pairs can be considered to a first approximation simply as a modification of the dielectric constant of the medium, this result indicates that the binding energy of the E_2H_2 exciton is modified very little (i.e., a decrease of 1 or 2 meV) since both the binding energy and the oscillator strength of an exciton display in general a similar dependence on the dielectric constant.

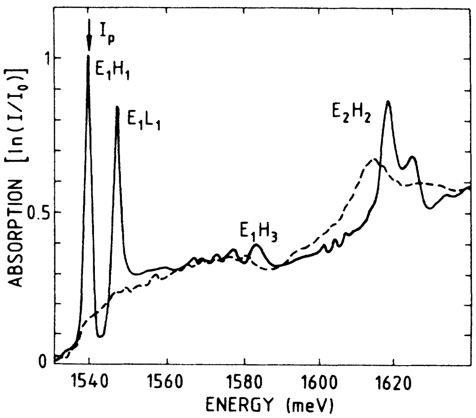

FIG. 1. Absorption spectra of unexcited (solid line) and excited (dashed line) GaAs/$Ga_{0.7}Al_{0.3}As$ MQW's (described in text). The pump (shown by arrow) is resonant with the E_1H_1 exciton and has an energy density of 80 μJ/cm^2 per pulse.

The observed red shift of the E_2H_2 excitonic resonance (2.1 meV) can therefore be assigned essentially to the BGR of the E_2 and H_2 subbands. This BGR arises from the exchange and correlation of a test E_2H_2 exciton (injected by the probe beam) with the dense population present in the first (E_1, H_1, and L_1) subbands. A word of caution is, however, necessary at this point: the shift measured in these spectra can only be taken as indicative of the order of magnitude of BGR for E_2H_2, which is of the order of a few meV for an average density of 3.5×10^{11} cm^{-2}. The reason that a more quantitative relationship between BGR and population density cannot be established is twofold: (1) Taking into account the slight decrease of the exciton binding energy increases the estimate for BGR by 1 or 2 meV. (2) The excitation of the sample is inhomogeneous. Even though the excitonic resonances of the first subband (E_1H_1 and E_1L_1) are completely bleached, the first few wells of the sample are probably excited to a higher density than the last few wells. The observed shift is thus a weighted average of the shifts in each individual well.

It is interesting to compare the order of magnitude of the BGR of E_2H_2 thus measured with the renormalization of the first subbands, which contain the dense population of carriers. However, the BGR of the first subbands cannot be observed directly in these absorption experiments because the spectral bleaching does not permit

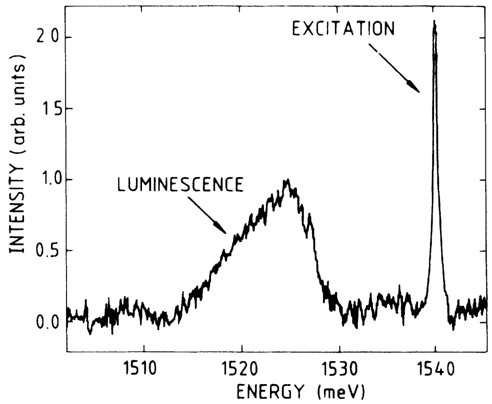

FIG. 2. Luminescence spectrum of the same MQW as in Fig. 1. Excitation is resonant with the E_1H_1 exciton, and has an energy density of 80 μJ/cm^2 per pulse.

us to localize the E_1H_1 band edge. However, information on the occupied states of the electron-hole plasma is obtained easily through luminescence experiments. The luminescence of this sample under the same excitation conditions (excited at the edge of the sample and observed at right angles with respect to the exciting beam) displays an emission peaked 12 meV to the red of the E_1H_1 exciton; that is, 20 meV to the red of the unexcited band gap, as seen in Fig. 2. The luminescence spectrum extends to about 30 meV to the red of the band gap, while its high-energy end is probably subject to a slight absorption as the light propagates a few tens of micrometers along the MQW layers, to reach the edge of the sample where it is detected. This may distort the exact shape of the luminescence peak, but nevertheless permits us to localize the low-energy end of the luminescence spectrum to approximately 30 meV below the (unexcited) band gap. A similar luminescence spectrum is obtained when the excitation beam (of the same intensity) is tuned to higher energies, in the band-to-band transition. On the other hand, when the sample is excited by a low-intensity beam (e.g., by a cw HeNe laser), its luminescence is peaked exactly on the E_1H_1 exciton. This result, as well as similar results in luminescence reported in the literature,[5,13] are in accord with the theoretical expectation for the extent of renormalization of a single occupied subband,[1] and indicates that the BGR is of the order of a few tens of meV in the E_1, H_1, and L_1 subbands, which are occupied.

These results show that BGR for an unoccupied subband is much smaller (by an order of magnitude) than the BGR of a subband that contains a large population of carriers. This difference goes against the naive model of BGR as a rigid shift of the whole band structure towards lower energies, but can be understood if we consider the exchange contribution to the BGR. A simple calculation of the exchange energy between two carriers (e.g., two electrons) in a quantum well of thickness L with infinite barriers height, each having a trial wave function of the form

$$\Psi(\rho,z)=\sin\left[\frac{n\pi z}{L}\right]e^{ik\rho}\,,$$

where n is the subband index, k is the in-plane wave vector, and ρ and z are cylindrical coordinates, gives

$$\epsilon\propto\frac{1\pm e^{-k_{12}L}}{(k_{12}L)^2+(n_1-n_2)^2\pi^2}+\frac{1\pm e^{-k_{12}L}}{(k_{12}L)^2+(n_1+n_2)^2\pi^2}\,,$$

where the indices 1 and 2 distinguish the two electrons, while $k_{12}=|k_1-k_2|$. Because of the term that depends on the difference of subband indices in the denominator of the first fraction the exchange interaction of carriers belonging to the same subband is much larger than for carriers belonging to different subbands. Thus, in a filled subband, the exchange interaction between carriers of the *same subband* gives rise to a large BGR. On the other hand, the exchange of a test carrier in an empty subband with the Fermi sea of a different subband changes very little the energy of the test carrier, and thus gives rise to a weak BGR. More sophisticated calculations on the subband dependence of the exchange energy have already appeared in the literature.[14]

In conclusion, in the experiments presented in this paper, we isolate the contribution of the many-body interactions in the excitonic optical nonlinearity of MQW by optically injecting E_1H_1 excitons and examining the changes that the injected species produce in the absorption spectrum of the E_2H_2 exciton. These spectral changes indicate that Coulombic screening is weak at the carrier densities attained (3.5×10^{11} cm^{-2}), while at the same time they provide a direct measurement of the quasi-2D band-gap renormalization. The magnitude of the renormalization of E_2H_2 obtained through nonlinear absorption techniques is smaller (by 1 order of magnitude) than the renormalization observed in luminescence experiments of the same sample under the same excitation conditions. This result indicates that the BGR due to a dense electron-hole–pair population is not the same for all states: it is large for the subbands that contain carriers, but relatively weak for unoccupied subbands. Thus, the description of BGR as a rigid shift of the whole band structure *is not* a good approximation, when states far above the Fermi level are under consideration.

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