Blue Shift of the Exciton Resonance due to Exciton-Exciton Interactions in a Multiple-Quantum-Well Structure

N. Peyghambarian, H. M. Gibbs, and J. L. Jewell[a]
Optical Sciences Center, University of Arizona, Tucson, Arizona 85721

and

A. Antonetti and A. Migus
Laboratoire d'Optique Appliquée, Ecole Polytechnique–Ecole Nationale Supérieure de Techniques Avancées, F-91120 Palaiseau, France

and

D. Hulin and A. Mysyrowicz
Groupe de Physique des Solides, Ecole Normale Superieure, F-75005 Paris, France
(Received 16 July 1984)

Femtosecond time-resolved dynamics of exciton screening and pairing is studied in a 53-Å GaAs multiple-quantum-well structure at 15 and 224 K. An intriguing high-energy shift of the exciton self-energy in the dense limit is reported.

PACS numbers: 71.35.+z, 46.20.+e, 78.20.−e

Multiple-quantum-well structures (MQWS) are currently attracting considerable interest, both from a fundamental point of view, as well as for their potential applications as efficient, active devices in optoelectronics.[1] They consist of alternate, ultrathin layers (typical thickness $d \simeq 100$ Å) of two semiconductors with different energy gaps. Carriers created in MQWS are confined in the layers of lower gap energy, thereby providing a rather unique quasi-two-dimensional system of electrons and holes. As a result of this confinement, the Coulomb attraction between electrons and holes is increased compared to that in bulk form of the same material. This leads to the observation of distinct exciton absorption lines in MQWS even at room temperature, which is very unusual for a normal semiconductor. Large optical nonlinearities, associated with the screening of these excitons upon optical creation of carriers, have been reported.[2] Based on these excitonic nonlinearities, room-temperature operation of all-optical devices (such as bistable elements) has been recently demonstrated.[3]

We have investigated the dynamics of the excitonic optical nonlinearities in a GaAs-AlGaAs MQWS, with an experimental apparatus having a time resolution of 120 fs. This study brings information about the dissociation rate of excitons into free electron-hole pairs, induced either by the presence of free carriers or by thermal phonons, as well as about the reverse effect, namely pairing of free carriers into excitons. We also report for the first time a renormalization of the exciton self-energy in a highly excited quasi-two-dimensional system. The principle of the measurement is similar to that pioneered by Shank *et al.*[4] An ultrafast optical pump pulse [duration full width at half maximum (FWHM) = 120 fs] creates a high density of electron-hole pairs in the GaAs layers of the sample. A weaker, broadband probe pulse, also of 120-fs duration, is transmitted through the excited sample at different delays from the time of carrier creation. By recording the pump-induced changes in the absorption spectrum of the probe pulse as a function of time delay, one has direct access to the evolution of the excited system. In the experiment both the pump and probe pulses are obtained by continuum generation in two different water cells using the amplified optical pulse from a ring cavity passively mode-locked (CPM) dye laser.[5] An interference filter selects a narrow spectral range in one of the continua for the pump pulse. Its pulse energy content is further amplified, if necessary, in a dye amplifier system.[6] By tilting of the interference filter, the pump peak wavelength may be finely adjusted so that the electron-hole pairs can be created either in the form of free carriers or directly as excitons. The sample itself consists of 300 periods of alternate layers of GaAs (thickness = 53 Å) and $Al_{0.3}Ga_{0.7}As$ (thickness = 56 Å) held at 15 K or 224 K. The time-resolved transmission spectra of the probe pulse are recorded with a spectrograph and optical multichannel analyzer.

Figure 1(a) shows spectra obtained at a sample temperature $T = 15$ K with the 740-nm wavelength of the pump pulse in the GaAs absorption continu-

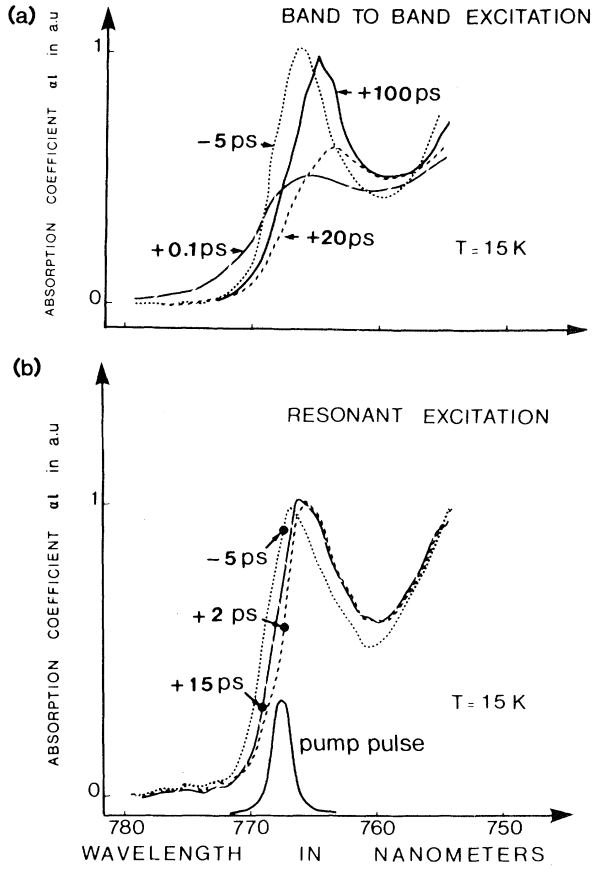

FIG. 1. (a) Time-resolved absorption of GaAs-$Al_{0.3}Ga_{0.7}As$ MQWS (GaAs layer thickness = 53Å) at $T = 15$ K recorded at various delays from the creation of free electron-hole pairs. The excitation wavelength is at 740 nm. (b) Excitation wavelength is adjusted at 767.7 nm inside the heavy-hole exciton line. The excitation laser spectrum is also shown.

um. The curve labeled -5 ps corresponds to a case where the probe pulse precedes the pump on the sample, and it is therefore representative of the unexcited crystal. The observed absorption line at 766 nm corresponds to the heavy-hole exciton. Immediately following the creation of hot carriers, one notices a strong modification of the absorption spectrum. The carriers screen the Coulomb interaction responsible for binding an electron and a hole to form a pair, leading to the disappearance of the exciton structures in the spectrum. We found that exciton bleaching requires a minimum pump energy of the order of 1 $\mu J/cm^2$ (corresponding to an estimated initial carrier density of 10^{17} cm^{-3}) and that it is accompanied by the appearance of a broad luminescence band characteristic of a two-component plasma in good agreement with similar results obtained by Shank *et al.*[4] In the results of Fig. 1, the pump energy is adjusted just above the threshold energy for plasma formation and the spectra are displayed at various delays. The time necessary to screen the exciton features is less than 150 fs. This is in agreement with similar experiments by Shank *et al.*[4] in GaAs where exciton screening was found to occur in less than 1 ps. However, the subsequent evolution of the system is markedly different. Whereas in bulk GaAs, complete exciton bleaching persists for delays in excess of several hundreds of picoseconds, here the exciton oscillator strength recovers rapidly, returning to its original value within 100 ps, an indication that the free carriers responsible for exciton bleaching have disappeared. Another striking feature is the high-energy shift of the peak of the exciton absorption as it reappears. Such a blue shift cannot arise from a pump-induced thermal effect that would instead displace the exciton toward lower energies. Rather, we attribute it to the fact that the sample is still in an electronically highly excited state, but with the electron-hole pairs in the form of a dense gas of excitons, rather than free carriers as initially injected. The blue shift then results from the mutual exciton-exciton interactions in the dense limit, leading to a renormalization of the exciton self-energy.

If this interpretation is correct, it should also be possible to observe situations where an exciton renormalization occurs instantly upon electron-hole pair creation, but without initial reduction of exciton oscillator strength, simply by adjusting the pump pulse wavelength inside the exciton resonance, in order to generate directly a high-density exciton gas. We have taken special precautions to obtain a narrow pump frequency spectrum, comparable in width to the heavy-hole exciton resonance.[7] This ensures that no significant density of free carriers is produced by a high-energy tail of the pump spectrum overlapping the band absorption continuum. The results of resonant pumping ($\lambda_{pump} = 767.7$ nm) are shown in Fig. 1(b). Consistent with the above model a blue shift without exciton bleaching is observed after exciton creation.[8] Delays in excess of 400 ps are necessary for the exciton resonance to gradually resume its normal, unperturbed position. This indicates that the exciton lifetime in this MQWS at 15 K is in the nanosecond range. The magnitude of the shift of the undiminished exciton energy is directly proportional to the pump intensity, i.e., to the exciton density.[9]

Another stringent test for this interpretation is obtained by performing similar experiments at temperatures close to the ambient. It has been deduced

by Chemla *et al.*[2] from a linewidth analysis that despite its pronounced appearance in absorption, the exciton lifetime in a MQWS at 300 K is 0.4 ps, due to thermal dissociation into a free electron and hole. It means that under nonresonant pumping, exciton bleaching should occur, but no shift, even at longer delays, since most of the generated electron-hole pairs always remain in the form of free carriers. On the other hand, under resonant pumping, an *initial excess population* of excitons is created. This should lead to a blue shift, together with bleaching, during the exciton lifetime, and thereby provide a means to verify directly the exciton thermal dissociation rate. Results obtained at $T = 224$ K are in agreement with this model as shown in Fig. 2. In both cases, at longer delays $\Delta t > 2$ ps, the behavior is the same, independent of the initial pumping conditions. A complete exciton screening is achieved, signaling the presence of long-lived carriers. The early behavior in the first picosecond is different, however, showing just bleaching in the nonresonant condition but simultaneous shifting and bleaching in the resonant pumping condition, as expected. Although a precise value of exciton lifetime is difficult to extract from a deconvolution of shift and bleaching, it is consistent with a value of 0.8 ps inferred from the difference in exciton linewidth, using the procedure of Chemla *et al.* for 224 K.

To further prove that the blue shift results from exciton-exciton interactions but not from "normal" (free carrier) screening, we show in more detail the behavior of the exciton resonance in the presence of an increasing density of free carriers (Fig. 3). This is obtained either by increasing the nonresonant pump intensity at a fixed, zero time delay ($T = 224$ K) or by recording the time evolution of the transmission spectrum during the pump pulse duration before the created carriers pair into excitons ($T = 15$ K). Note the absence of any shift in both cases, as the exciton peak absorption progressively decreases.

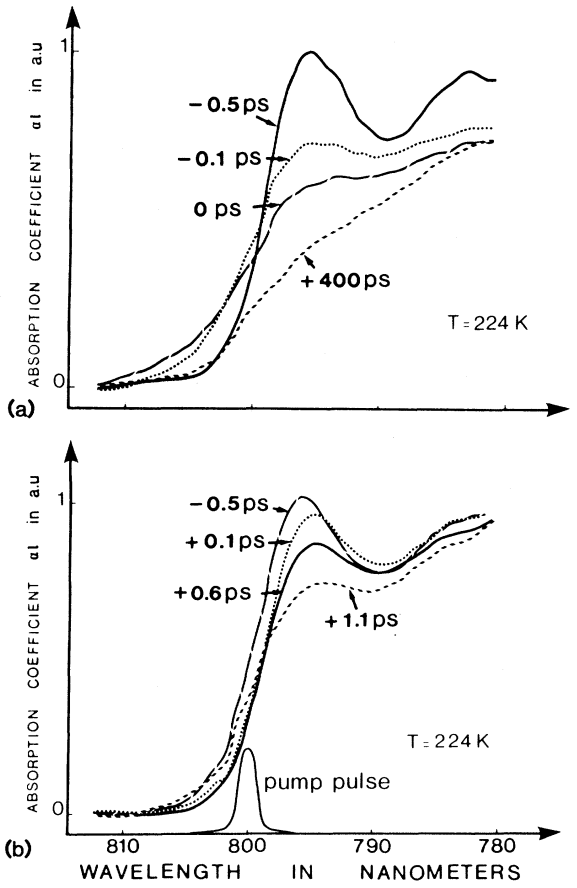

FIG. 2. Time-resolved absorption spectra of GaAs MQWS at $T = 224$ K at different delays from the excitation pulse. In (a) the pump pulse wavelength is at 760 nm (nonresonant excitation), while in (b) it is at 800 nm (resonant excitation).

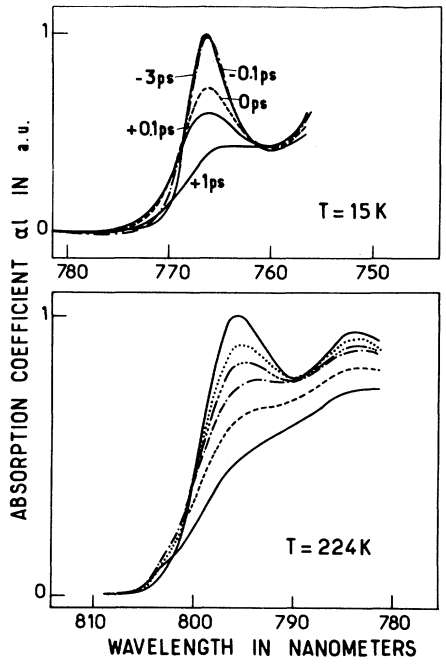

FIG. 3. Time-resolved absorption spectra of GaAs MQWS showing the onset of exciton bleaching under nonresonant pumping. The upper curve gives the time evolution of the exciton during the pump pulse, for a fixed pump intensity. The lower curve is obtained at fixed zero delay with a pump intensity increasing by successive steps of 2.

A renormalization of the exciton self-energy to higher values is not predicted in current theoretical models[10] that are applicable to three-dimensional crystals. Recently, Fehrenbach *et al.*[11] have performed an experiment in bulk GaAs, in which excitons are resonantly created at low temperature with a picosecond laser. No displacement of the exciton was observed, up to excitation intensities for which the excitonic gas underwent a transition into a plasma. This suggests that the shift observed here is related to the reduced dimensionality of the electron-hole system in the superlattice structure used here. To the best of our knowledge, there are no theoretical calculations of the exciton self-energy pertaining to the two-dimensional exciton in the dense limit.

In conclusion, we have observed an intriguing blue shift of the exciton resonance in a highly excited multiple-quantum-well structure, which we attribute to predominantly repulsive interactions between excitons in the dense regime. At elevated temperatures creation of a high density of nonequilibrium excitons results in a blue shift whose disappearance allows a direct measurement of their thermal dissociation time.

Part of the results of this work have been presented as a postdeadline paper at the Thirteenth International Quantum Electronics Conference, Anaheim, California, 1984. This work has been partially supported by a contract from the Direction des Recherches Etudes et Techniques, by the Centre National de la Recherche Scientifique, by the U.S. Air Force Office of Scientific Research, by the U.S. Army Research Office, and by the National Science Foundation. We thank A. C. Gossard and W. Wiegmann (AT&T Bell Laboratories, Murray Hill, N.J. 07974) for the growth of the MQW crystal. Groupe de Physique des Solides is a laboratoire associé au Centre National de la Recherche Scientifique.

---

[a]Presently at AT&T Bell Laboratories, Holmdel, N.J. 07733.

[1]L. Esaki and R. Tsu, IBM J. Res. Dev **14**, 61 (1970); R. Dingle, in *Festkorperprobleme: Advances in Solid State Physics,* edited by H. J. Queisser (Pergamon, Vieweg, Braunschweig, 1975), Vol. 15, p. 21; A. C. Gossard, Treatise Mat. Sci. Technol. **24**, 13 (1982).

[2]D. S. Chemla, D. A. B. Miller, P. W. Smith, A. C. Gossard, and W. Wiegmann, IEEE J. Quantum Electron. **20**, 265 (1984).

[3]H. M. Gibbs, S. S. Tarng, J. L. Jewell, D. A. Weinberger, K. Tai, A. C. Gossard, S. L. McCall, A. Passner, and W. Wiegmann, Appl. Phys. Lett. **41**, 221 (1982).

[4]C. V. Shank, R. L. Fork, R. F. Leheny, and J. Shah, Phys. Rev. Lett. **42**, 112 (1979); C. V. Shank, R. L. Fork, B. I. Greene, C. Weisbuch, and A. C. Gossard, Surf Sci. **113**, 108 (1982).

[5]R. L. Fork, B. I. Greene, and C. V. Shank, Appl. Phys. Lett. **38**, 671 (1979).

[6]A. Migus, J. L. Martin, R. Astier, A. Antonetti, and A. Orszag, in *Picosecond Phenomena III* (Springer Verlag, New York, 1982), p. 6.

[7]Pump pulse frequency narrowing inevitably leads to a deterioration of the temporal resolving power of the system, which remains, however, well in the subpicosecond domain (pump pulse duration after narrowing at 800 nm is 250 ps).

[8]We have verified that the shift is insensitive to the exact position of the pump beam within the line profile. During the first two picoseconds at low temperatures, in addition to the blue shift of the entire exciton resonance there is a small dip in the absorption at an energy below the pump energy, which remains to be studied.

[9]At still higher pump intensities, shifts similar in magnitude to those of Fig. 2(a) are observed. However, exciton bleaching sets in.

[10]R. Zimmermann, K. Killmann, W. A. Kraeft, D. Kremp, and G. Ropke, Phys. Status Solidi **90**, 175 (1978); J. G. Gay, Phys. Rev. B **4**, 2567 (1971).

[11]G. W. Fehrenbach, W. Schafer, G. Treusch, and R. G. Ulbrich, Phys. Rev. Lett. **49**, 1281 (1982).