Femtosecond Dynamics of Resonantly Excited Excitons in Room-Temperature GaAs Quantum Wells

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We investigate the effect of excess excitonic populations created by resonant ultrashort excitation on the optical-absorption properties of GaAs quantum wells. We find that under these conditions at room temperature excitons produce *more* absorption bleaching than equal densities of free-carrier pairs. This bleaching partly recovers as the excitons ionize to give free carriers. Hence, we directly measure the thermal ionization time of excitons at room temperature for the first time, and find that $\tau \sim 300$ fs.

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At low excitation levels, the optical-absorption spectrum near the direct band gap in pure semiconductors is dominated by optical transitions involving correlated electron-hole pairs, with clear exciton resonances existing in many such materials at low temperature. With increasing densities of free electrons and holes, the Coulomb interaction between electrons and holes (which causes the exciton binding) is screened. Furthermore, the conduction- and valence-band states near the band edge become occupied (this "band filling" of itself leads to absorption bleaching) and the band-gap energy renormalizes to lower energy. Such effects have been investigated both in bulk GaAs[1,2] and quasi-two-dimensional GaAs multiple-quantum-well structures (MQWS)[3] by use of ultrashort light pulses, of photon energy much greater than the band-gap energy, to excite the material; this technique creates free electron-hole pairs directly and allows the dynamics of the processes to be studied. The exciton resonances are readily screened in both cases.

By contrast, recent low-temperature investigations in GaAs[4] have shown that when excitons are created directly by exciting with a light pulse at the exciton absorption resonance, the screening of excitons by excitons is very weak compared to the screening of excitons by free carriers; the energy of the exciton peak changes very little even at large exciton densities. In the present paper we investigate the bleaching of the exciton resonance absorption by both free carriers and excitons, using GaAs MQWS at room temperature. We find that, although there is little shift of the exciton resonance with increasing densities, the bleaching effect of excitons on excitons can actually be *stronger* than that of the same density of free carriers. There is therefore a strong interaction between the excitons under our conditions. We perform this experiment by use of ultrashort light pulses. We are able to time resolve the ionization of excitons by optical phonons, and deduce a lifetime of ~ 300 fs for the excitons at room temperature.

MQWS at room temperature offer a unique system in which to study these effects. First, the MQWS is exceptional in that it shows clear exciton resonances at room temperature[5]; this is partly due to the increased binding energy of the confined excitons.[6] Second, the ionization of excitons by optical phonons to give free carriers is expected to be a strong process with an ~ 400-fs exciton lifetime predicted[7]; thus effectively all the excitons will be converted to free carriers, with competing exciton-decay processes being negligible. Hence in one time-resolved experiment we can directly compare the effects of the same density of excitons and free carriers. (We also test this with a control experiment in which we create free carriers directly by excitation above the band gap.) Third, the free carriers, whether created directly or through exciton ionization are "warm" and remain so.

The sample which we studied consists of 65 periods of GaAs quantum wells 96 Å thick, alternated with $Al_{0.3}Ga_{0.7}As$ barrier layers 98 Å thick. The MQWS was grown between two 1-μm cap layers of AlGaAs on a GaAs substrate by molecular-beam epitaxy. The substrate was removed by selective etching over part of the sample and antireflection coatings of silicon oxide were deposited on both faces to effectively eliminate Fabry-Perot effects. The resulting sample had sufficiently good optical quality that pump-beam scattering was insignificant.

The experiments were performed using femtosecond optical measurement techniques.[8] Optical pump and probe pulses were obtained by amplifying pulses from a colliding pulse laser at 620 nm at a repetition rate of 10 Hz[9] and then focusing those pulses into a 1-mm-thick ethylene glycol jet to form broadband continuum pulses. One portion of the continuum was separated and used as a probe. The other part

was amplified[10] in a 1-cm cell of LDS 821 dye in propylene carbonate solvent pumped by 30 mJ of 532-nm light. A pair of diffraction gratings removed group-velocity dispersion from the amplified pulses, a spatial filter rejected amplified spontaneous emission, and interference filters of 10-nm bandwidth selected the desired pump-pulse spectrum with energies up to 50 nJ. The amplified infrared pulses were 150 fs in duration and set the limiting time resolution. This apparatus allowed us to probe the infrared absorption spectrum of the MQWS while pumping with a continuously tunable infrared pulses.

The absorption spectrum of the unperturbed sample [Fig. 1(a)] has been extensively discussed.[6,7] We used two different pumping wavelengths, one "resonant," designed to create primarily (heavy-hole) excitons, and the other "nonresonant" to create primarily free carriers. For resonant pumping at room temperature we used a distribution centered at ~ 855 nm (1.45 eV) and for nonresonant pumping a distribution centered at ~ 812 nm (1.53 eV) [Fig. 1(d)]. The absorption spectra under resonant pumping at a density of 7×10^{10}-cm^{-2} excitons per layer[11] are shown in Fig. 1 for three times shortly after excitation with the unperturbed spectrum as a reference. We used this density because the absorption changes are large enough to be easily measured while the excitation peaks are still resolved. The resolved peaks indicate that excitons still exist at this density. We observe a negligible shift of the exciton peak (less than 1 meV) at this excitation density.

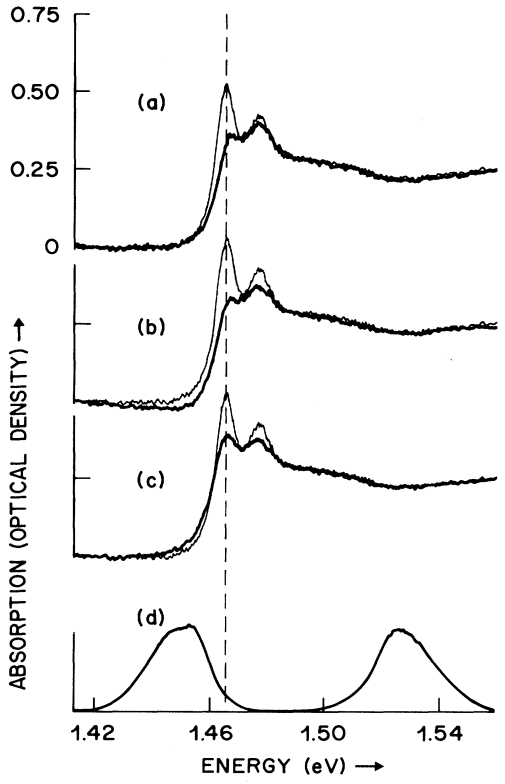

FIG. 1. Comparison of the unpumped MQWS sample absorption spectrum to the spectra under resonant excitation at various delays between the pump and the probe continuum: (a) at the peak of the excitation pulse, (b) 100 fs after the peak, and (c) 500 fs after the peak. In (d) the spectra of the resonant and nonresonant pump pulses shown. The dashed line shows the position of the heavy-hole exciton peak.

We find two time regimes for the case of resonant pumping at room temperature. For times < 500 fs we observe rapid changes in the absorption spectrum. For times > 500 fs the spectra change much more slowly indicating that a quasiequilibrium condition has been reached. In this quasiequilibrium regime, the absorption spectrum is not sensitive to the pump wavelength. With resonant pumping we observe an efficient bleaching of the heavy-hole exciton peak which partially recovers within less than 1 ps (Fig. 1). This behavior is observed with pump and probe pulses generated in the same continuum jet, in separate jets, and for both parallel and orthogonal pump and probe polarizations. It occurs, however, only with resonant pumping. In contrast, with nonresonant pumping, we observe a progressive bleaching of the exciton feature with an immediate increase in absorption below the heavy-hole exciton peak similar to that reported previously.[7]

In Fig. 2 we compare the time dependence of the absorption at the peak of the heavy-hole exciton for

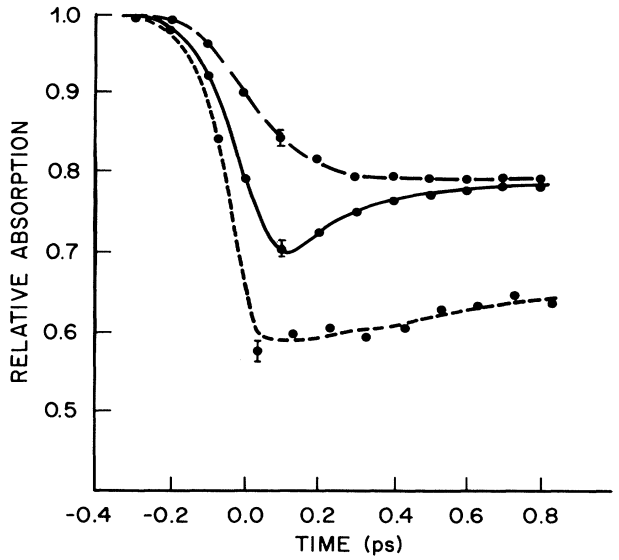

FIG. 2. Dynamics of the changes of the heavy-hole exciton peak absorption as a function of the pump-probe delay. Dashed line, nonresonant, 300 K; solid line, resonant, 300 K; and dot-dashed line, resonant, 15 K.

resonant and nonresonant pumping. We have adjusted the relative intensities in the two pumping cases so that the spectra of the excited sample are identical for times greater than 1 ps. In so doing, we ensure that the excited pair density is essentially equal in the two pump cases, thus making a detailed comparison possible. This figure shows the dramatic difference in the dynamics of the heavy-hole exciton peak absorption created by the two pump conditions. In the nonresonant case, the absorption bleaching closely follows the integral of the instrumental response (<100 fs response time), as expected due to Coulomb screening. In the resonant case a transient nonequilibrium situation is clearly observed. This behavior can only be explained by assuming that the initially excited excitons bleach the excitonic absorption more efficiently than an equal density of carrier pairs excited at ~ 300 K. Using a simple rate-equation analysis, we deduce that the excitons produce approximately *twice* the absorption bleaching of an equal density of uncorrelated carrier pairs. We also obtain an ionization time of 300 ± 100 fs, in good agreement with line-shape predictions[7] and the mean time for collisions between LO phonons and electron-hole (e-h) pairs.[12] The mechanism that emerges which is consistent with our data is that the initially excited excitons bleach the excitonic absorption and as the excitons become ionized (by thermal phonons), the absorption returns to the plateau value determined by Coulomb screening of e-h pairs with a ~ 300-K temperature.

We tested the role of thermal phonons by repeating the resonant-pumping experiment at a temperature of 15 K where the thermal-LO-phonon density is negligible. In this case we observe a similar bleaching of the exciton resonance and negligible spectral shift. The recovery time is, however, considerably longer (~ 10 ps) (Fig. 2). The absence of a prompt recovery at low temperature is consistent with exciton ionization via phonon interactions as the source of the rapid absorption recovery at room temperature.

The question then arises as to the mechanism by which excitons bleach excitons. We believe that the mechanism is excitonic phase-space filling. Excitonic states are built up from single-particle states of the crystal and thus the resonant generation of excitons produces a phase-space filling which is at the origin of the decrease of the exciton absorption. This mechanism has been invoked semiempirically by several authors[7,13] and recently a more rigorous theoretical description of exciton-exciton interactions based on many-body theory has been developed.[14] The mechanism in its simplest form consists of reducing the excitonic optical absorption in proportion as the space (either real space or phase space) is filled with excitons. To make the model quantitative the theory must calculate the effective area of the exciton. With use of the many-body formalism, it can be shown[15] that, to first order in the exciton density per unit area N, the absorption can be written as $\alpha=\alpha_0(1-N/N_s)$. In this expression, the saturation parameter $1/N_s$ has dimensions of area (i.e., the effective exciton area) and in the case of phase-space filling by quasi-two-dimensional excitons can be expressed as $1/N_s = 32\pi a_0^2/7$ where a_0 is the Bohr radius. From the experimental data, we deduce an effective cross section (change of absorption per exciton) of 10^{-13} cm^2 which corresponds to $a_0 = 55$ Å, in excellent agreement with the value $a_0 = 64$ Å calculated using a variational procedure for our sample parameters.

Therefore, our explanation for our observation that excitons produce more absorption bleaching than an equal density of free e-h pairs is as follows. Resonant excitation creates excitons which have no excess energy (equivalent to $T=0$ K) before interacting with the lattice. The ionization of the excitons, through interaction with thermal LO phonons, produces e-h pairs that are at a much higher temperature since each carrier pair receives ~ 25 meV of excess energy from a LO-phonon collision. The phase-space filling due to excitons is large, whereas the band filling and screening due to free carriers are strongly reduced at high temperature.

This bleaching of the excitonic absorption by excitons should not be peculiar to the MQWS. Indeed, the low-temperature GaAs results[4] do show bleaching under resonant pumping. However, it is not currently clear either theoretically or experimentally whether the relative strength of exciton and free-carrier bleaching effects is the same in bulk material. There is some theoretical evidence that screening is weaker in the quasi-two-dimensional system,[16] but the detailed physics of screening in both confined and bulk systems is still unclear.

In conclusion, we observe for the first time saturation of the (heavy-hole) exciton peak in MQWS by the direct creation of excitons. Subsequently, through ionization of the excitons on a femtosecond time scale, free e-h pairs are generated which then produce a Coulomb screening of the excitonic absorption. The saturation which we attribute to this Coulomb screening is experimentally the same as that produced by direct excitation of free e-h pairs while the saturation due to the directly created excitons is qualitatively different. All the absorption changes associated with these processes occur within 500 fs. We deduce from our measurements an exciton ionization time of 300 ± 100 fs which is consistent with previous estimates for optical-phonon ionization. In addition, our results suggest that at room temperature the effect of ultrafast selective generation of excitons on the absorption spectrum can be greater than that of the same density of free e-h pairs, despite the reduced Coulomb

screening in the exciton case.

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