Femtosecond Excitation of Nonthermal Carrier Populations in GaAs Quantum Wells

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We report the first direct observation of nonthermal photoexcited carrier distributions in GaAs quantum-well structures. We present experimental studies which show that these distributions thermalize within 200 fs. In addition we are able to show that near the band edge the effect of long-range Coulomb screening on the bleaching of the two-dimensional-exciton resonances is much weaker than that of the Pauli exclusion principle.

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Ultrashort-optical-pulse techniques have been used to excite hot carriers in semiconductor multiple-quantum-well structures and to probe the process of carrier cooling to the lattice temperature.[1,2] In the experiments reported here we excite a nonequilibrium carrier population in such a short time that the temperature of the distribution cannot yet be defined. Using femtosecond spectroscopic techniques we are able to follow the evolution of the nonthermal carrier distribution to a thermalized distribution within 200 fs at room temperature in a GaAs multiple-quantum-well structure (MQWS). By monitoring the time development of the bleaching of the exciton resonance we are able to determine for the first time the relative effectiveness of Coulomb screening and Pauli exclusion on quasi two-dimensional exciton absorption.

Optical transitions in a semiconductor are strongly influenced by the distribution of carriers in the conduction and valence bands. For direct transitions away from exciton resonances the absorption coefficient can be expressed as

$$\alpha = \alpha_0(1 - f_e - f_h),$$

where $f_{e,h}$ are the distribution functions for electrons and holes and α_0 is the absorption coefficient in the absence of state filling. With a sufficiently short optical pulse a narrow band of states can be excited, creating a nonthermal distribution. As time progresses, carrier-carrier interactions lead to thermalization of the carrier distribution. Ultimately, at much longer times the carriers equilibrate with the lattice.[1,2] In direct–band-gap semiconductors carrier-carrier scattering is extremely fast and only a very limited number of experimental studies of this process have been reported. They include a single-wavelength nonlinear polarization-rotation measurement at low temperature in bulk GaAs[3] and a single-wavelength nonlinear transmission measurement high above the band gap of AlGaAs.[4] However, by use of femtosecond spectroscopic techniques[5–7] which have been recently extended to kilohertz repetition rates,[8] the dynamics of nonthermal carrier distributions can be directly monitored by measurement of the time development of the absorbance near the pumping energy.

For optical transitions near an exciton resonance the influence of carriers is more complex.[9] Carriers can modify the excitonic absorption through the effects of the exclusion principle or through Coulomb screening. In this work we resolve these two mechanisms for the first time. In quasi two-dimensional semiconductors, exciton resonances are well resolved at room temperature and have been the subject of extensive experimental[10] and theoretical work.[11,12] In previously reported work[13] femtosecond optical pulses were used to excite excitons resonantly in a room-temperature GaAs MQWS and the exciton ionization time was measured in a room-temperature phonon bath by monitoring of the bleaching of the exciton resonance. In a recent theoretical study, Schmitt-Rink, Chemla, and Miller[12] have determined the influence of carriers excited above the band edge on the exciton resonance absorption. In their study they assess the relative effectiveness of excited carriers on exciton absorbance through direct Coulomb screening and through Pauli exclusion, i.e., the effect of occupation of near–band-edge states, which they refer to as phase-space filling and exchange screening.

In the experiments reported here we monitor the change in exciton absorbance early in time induced by the nonthermal population distribution and we follow the absorbance changes as the carriers thermalize and subsequently occupy near–band-edge states. This allows us to determine for the first time the relative importance of these two processes on bleaching the exciton resonance. All these measurements were performed at room temperature.

Using a high-repetition-rate (8 kHz) data acquisition system we have been able to measure time-resolved spectra of induced transmittance changes to an accuracy of one part in 10^4 with better than 100-fs time resolution. Optical pulses of 50-fs duration were generated in a colliding-pulse mode-locked laser[5,7] and amplified to ~ 3-μJ energies at 8-kHz repetition rate with a copper-vapor-laser–pumped amplifier system.[8] These pulses are used to generate white-light continuum pulses. About 5% of the continuum beam was used as

a broadband 50-fs probe pulse to measure the absorption of the sample over a 150-meV portion of the spectrum, covering both the $n=1$ and the $n=2$ resonances. The remainder of the beam was passed through an interference filter with a 10-nm bandwidth. The pump pulse was broadened to approximately 100 fs as a result of the spectral narrowing upon passage through the filter. This spectral filtering is not ideal, and may produce small wings on the pump pulse. The pump and probe beams were focused to a ~ 20-μm-diam spot. A stepping motor was used to provide the relative time delay and the spectra were measured in the differential transmittance mode of an optical multichannel analyzer (OMA III) with a reticon detector. In such a mode, the data acquisition system measures directly the transmittance difference between the excited and the unexcited samples normalized to that of the unperturbed sample, i.e., $(T_0-T)/T_0$, which for small changes of the transmission is proportional to $\alpha_0-\alpha$. In the following we refer to these spectra as differential transmittance spectra (DTS). The sample is a MQWS that consists of 65 periods of 96-Å GaAs quantum wells and 98-Å $Al_{0.3}Ga_{0.7}As$ barrier layers. This sample has been described and analyzed by Chemla *et al.*[14]

In Fig. 1 we have plotted three DTS taken at nominally zero relative time delay for three pump-photon energies. An increase in the transmittance is seen in the energy range near the pump energy, which is indicated by an arrow for each case. The magnitude of the transmission change at the exciton[14] yields an estimated excitation density of 2×10^{10} cm^{-2}. A simple calculation, in the effective-mass approximation,[15] shows that at this carrier density a transmission change of $\sim 1.3\%$ is expected in the region of the pump spectrum. We observe a peak signal of $\sim 0.5\%$, which is consistent with our estimate. The dependence of the differential transmittance spectra on the pump energy is clear evidence of a nonthermal carrier distribution occupying states excited by the tunable pump pulses. Also observed is a small increase in transmission near both the $n=1$ and the $n=2$ excitons. The DTS profile close to the excitons is characteristic of broadening of resonances and reduction of oscillator strength.[10] At these early times the nonthermal carriers occupy states of the $n=1$ continuum which do not enter in the linear combination from which the $n=1$ excitons are constructed and do not belong to the $n=2$ subband states. Thus they only produce a weak Debye screening on the exciton resonances and there is no effect due to the Pauli principle. The Debye screening, of course, does not depend strongly on the exact energy distribution of the carriers. The relative absorption changes at $n=2$ are smaller than at $n=1$ because the $n=2$ excitons have a smaller oscillator strength and a broader resonance.

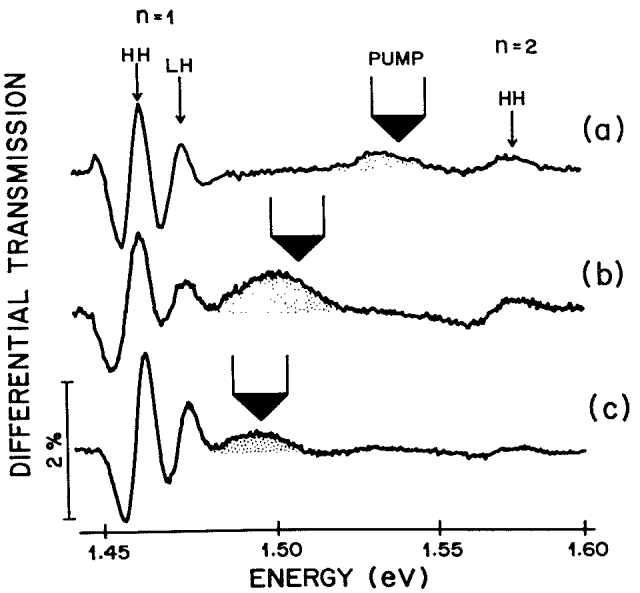

FIG. 1. Differential transmission spectra at nominally zero delay. The three spectra correspond to excitation by 100-fs pulses centered at (a) 1.537 eV, (b) 1.509 eV, and (c) 1.496 eV. The spectral width of the exciting pulse is ~ 20 meV. HH and LH refer to heavy-hole and light-hole excitons, respectively.

The temporal evolution of the nonthermal population is presented in Fig. 2. DTS at intervals of 50 fs for delays varying between -100 and 200 fs are shown. The point of zero time delay is estimated with a precision of approximately 50 fs. The pump energy has a distribution 20 meV wide and is centered at 1.509 eV as shown by the spectrum at the bottom of the figure. In the first spectrum, a well-defined region of increased transmission is observed which is broader than the pump spectrum and slightly shifted to lower energies. As time evolves the region of higher transmission shifts to lower energy and broadens. As the carriers begin to thermalize, near–band-edge states in the $n=1$ continuum become occupied and phase-space filling and exchange bleach the $n=1$ exciton absorption more efficiently. It is important to note that the bleaching of the $n=2$ exciton remains essentially constant as the carriers thermalize because for these resonances the only bleaching mechanism is the (weak) long-range Coulomb screening. Let us stress that the absorbance changes at the $n=2$ excitons serve as an internal probe of the pure Coulomb screening and show that it does not depend strongly on the carrier distribution. The increase in transmission near the $n=1$ excitons is delayed in time compared to the absorbance changes near the pump energy, showing that the exciton bleaching does not reach its peak value until the carriers thermalize and fill the near–band-edge states.

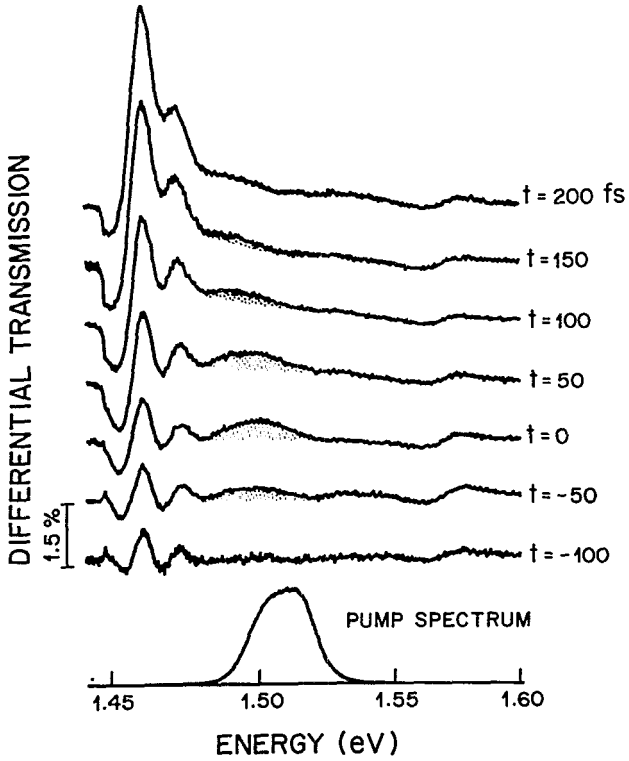

FIG. 2. Differential transmission spectra for a pump centered at 1.509 eV and observed at 50 fs intervals between $t = -100$ fs and $t = +200$ fs. The spectra have been displaced vertically for clarity. The pump spectrum is shown at the bottom of the figure. Various features are discussed in the text.

After 200 fs the spectra remain practically unchanged showing that the carriers have thermalized. We note that the carrier temperature at this time may not yet be the same as the lattice temperature. This final equilibration will take place on a picosecond time scale.[1,2] By numerical integration of the DTS above the $n = 1$ resonances, we estimate that the average carrier energy remains essentially constant during the thermalization process, indicating very little interaction with the lattice. This is as expected, since we excite carrier pairs with less than one LO-phonon excess energy, making LO-phonon emission forbidden, and the carrier-phonon collision time for LO-phonon absorption is approximately 300 fs under these conditions.[14] Therefore carrier-carrier scattering must be the dominant thermalization mechanism in this experiment. The initial evolution of the continuum-state bleaching indicates that the carriers leave their initial states with a response time of 50–100 fs, and the exciton bleaching appears with a delay time of approximately 150 fs. From the data we estimate that exciton bleaching due to screening is at least six times smaller than that due to phase-space filling and exchange in accord with the recent predictions.[12]

In conclusion, we have excited a nonthermal population distribution in a GaAs multiple-quantum-well structure and probed its evolution to thermalization. Since the carriers were excited with excess energy of less than one LO-phonon energy, no significant energy was exchanged with the lattice. The excited carriers were observed to interact rapidly to form a thermalized distribution within 200 fs. The simultaneous measurement of the absorbance changes at the $n = 1$ and the $n = 2$ exciton resonances gives the first direct evidence that Pauli-exclusion effects dominate over Coulomb screening in bleaching the exciton absorption in MQWS at room temperature.

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