Femtosecond Circular Dichroism Study of Nonthermal Carrier Distributions in Two- and Zero-Dimensional Semiconductors

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We report the time-resolved circular dichroism induced by nonthermal distributions of carriers in GaAs quantum wells, separating completely, for the first time, the effects of screening and phase space filling. Thermalization of the initial excitation is strongly inhibited when carriers are laterally confined into zero dimensions by a large perpendicular magnetic field.

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The linear and nonlinear optical properties of semiconductors depend upon the dimensionality of the materials [1]. Recently, nonlinear optical studies of quantum wells subjected to a perpendicular magnetic field have demonstrated that zero-dimensional magnetic confinement strongly reduces the Coulomb interactions of excitons created using either linearly or circularly polarized photons [2,3]. The dramatic effects associated with confinement to zero dimensions (0D) are not restricted to the electron-hole (*e-h*) pairs correlated into the discrete excitonic states; electron-hole pairs excited above the band gap into two-dimensional (2D) scattering states exhibit a remarkable transformation in their dynamical processes when they are confined into 0D. Previously [4], time-resolved measurements have demonstrated that the nonlinear optical response during the thermalization of an excited 2D plasma is dominated by a combination of screening and phase space filling (PSF). In this Letter, we report the first measurements of the circular dichroism (CD) response of a nonthermal distribution of 2D carriers. These time-resolved CD measurements enable a complete separation of the two principal components of the nonlinear optical response: screening and phase space filling. The occupation of angular momentum states by the excited carriers is directly observed in these measurements, allowing a new and detailed picture of the changing occupation of these states during the process of thermalization. Repeating these measurements in a 12-T perpendicular magnetic field demonstrates the effects of 0D confinement on the screening, PSF, and thermalization processes.

Nonlinear optical measurements are performed in the excite-and-probe transmission geometry, in which a strong tunable pump pulse excites the sample, and a weak, broadband probe pulse measures the absorption of the excited material. The sample studied is an 8.4-nm multiple quantum well structure, in the intrinsic region of an unbiased *p-i-n* diode. All measurements were performed at 4 K. To facilitate these measurements, optical pulses, with a center wavelength of 805 nm and approximately 100 fs duration, are generated by a synchronously pumped dye laser, and amplified in a copper vapor laser pumped dye amplifier to produce pulses with energies of 1 μJ, at a repetition rate of 8 kHz [5]. These pulses are focused into a chirp-compensated continuum generator, to produce broadband light pulses, which retain their short pulse width and have spectral components extending from the infrared to the ultraviolet. This continuum beam is split to produce the strong pump and weak probe beams, with the pump filtered spectrally and delayed relative to the probe. Both beams are independently circularly polarized, and focused onto the GaAs quantum well sample, located in the center of a 12-T superconducting magnet. After passing through the sample, the probe beam is collected into an optical fiber for parallel detection using an optical multichannel analyzer.

The spectrum of the pump is filtered to produce a pump excitation approximately 25 meV above the lowest heavy-hole (1*s* hh) exciton, with a bandwidth of 10 meV. Photons absorbed from this pump pulse produce electron-hole pairs with a distribution determined by the pump spectrum. The electron and hole created during the absorption of the pump photon each carry off excess kinetic energy according to the ratio of their masses. Heavy holes acquire a kinetic energy of 3 meV, while their associated electrons acquire 22 meV; light holes (lh), with a confinement energy 11 meV larger than that of the heavy holes, carry off 5 meV, with their associated electrons gaining 9 meV. These carriers separate, interact with phonons and other photoexcited carriers, and lose coherence with each other. The nonlinear optical response of the quantum wells results from the effects of this photoexcited *e-h* plasma upon the optical properties as measured by the probe.

The nonlinear optical response for identical pump and probe polarizations [Fig. 1(a)] exhibits the phase space filling and screening effects associated with the optical excitation of approximately 10^{10}-cm^{-2} carriers. When the probe arrives before the pump ($\Delta t = -400$ fs) the nonlinear response is very small. When the probe and pump are coincident in the material ($\Delta t = 0$ fs), a spectral hole is produced in the scattering state absorption due to the nonthermal distribution of occupied scattering states. The spectral hole appears because electron and hole states become occupied upon the absorption of a pump photon; their phase space is filled, and therefore unavail-

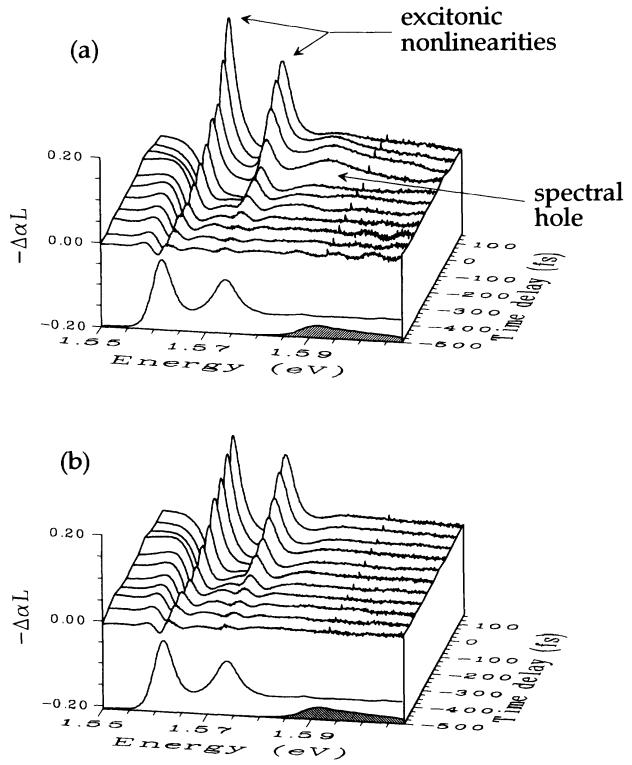

FIG. 1. Spectral hole-burning signal at zero field, for (a) identical and (b) opposite circularly polarized pump and probe. The linear absorption (solid) and pump spectrum (hatched) are shown in the foreground.

able for the subsequent absorption of a probe photon. This broad peak in the induced transparency ($-\Delta\alpha L$) is accompanied by a relatively large signal at the $1s$ hh and $1s$ lh excitons (linear absorption and pump spectrum, Fig. 1 foreground). These large signals arise from screening of the excitons. The signal at the heavy-hole exciton shows increased transmission on the high-energy side of the resonance, with decreasing transmission below, indicating a weak red shift of the exciton, in agreement with theoretical predictions [6]. As time progresses, and the pump is absorbed in the material, the nonthermal distribution experiences strong scattering that thermalizes it at an elevated temperature. Heavy holes, light holes, and conduction electrons each thermalize to the bottom of their respective band. The spectral hole burned into the nonlinear spectrum therefore evolves to a smooth nonlinear signal that decreases monotonically from the ionization threshold out to higher energies. The signal at the excitons rises steeply until the pump is absorbed, and thermalization complete.

The nonlinear response for oppositely polarized pump and probe [Fig. 1(b)] shows the same screening behavior at the excitons, with the signal rising during the absorption of the pump pulse and thermalization of the photoexcited plasma. The spectal hole-burning signal in the scattering states, however, is completely absent. To understand this, note that pump photons with $\sigma-$ polarization excite electrons from the $m_j=\frac{3}{2}$ heavy valence band into the $m_j=\frac{1}{2}$ conduction band; electrons from the $m_j=\frac{1}{2}$ light valence band are excited into the $m_j=-\frac{1}{2}$ conduction band. Probe photons behave similarly, the signs of all angular momentum projections being reversed in the case of $\sigma+$ polarized probe photons. Measurements using counterpolarized pump and probe therefore differ from copolarized measurements in the angular momentum of the electrons and holes measured by the probe. This affects the measured nonlinear optical response since screening is independent of angular momentum, while PSF is not. Because an oppositely polarized probe couples heavy holes to those conduction band states occupied by electrons pumped from the light-hole band, and vice versa, some small PSF signal is expected even in the case of counterpolarized pump and probe. The magnitude of this signal, however, is small and energetically separated from the observed hole-burning signal for copolarized pump and probe; it is not resolved in these measurements [7]. The measured counterpolarized nonlinear response therefore results entirely from (spin-independent) screening. Since the thermalization time is comparable to the pulse width, the rise in exciton screening signal is a combination of the increasing density of excited carriers, which follows the time integral of the pulse width, and the changing efficiency of screening as the carriers thermalize. Furthermore, the absence of the transient spectral hole and subsequent thermalized absorption bleaching, in the nonlinear response for counterpolarized excitation, demonstrates that the initially spin-polarized angular momentum distribution is not randomized during the process of thermalization, in agreement with previous experiments [8].

The separation of the pure screening component of the nonlinear optical response, via the measured nonlinearity under oppositely polarized pump and probe, enables a further analysis of the contribution from PSF effects. Assuming that charge-density effects dominate the nonlinear response for counterpolarized excitation, the measured counterpolarized response may be subtracted from the full nonlinear response for the copolarized condition, to yield the CD response resulting purely from the PSF component of the nonlinearity. When this is done (Fig. 2), the evolution of the charge distribution, before and after thermalization, is readily apparent. When pump and probe coincide, $\Delta t=0$ fs (solid), the spectral hole in the scattering states has the background effects of screening removed, and is clearly resolved before thermalization. No linearity is observed at the heavy-hole excitons for $\Delta t=0$ fs, because the excitons are formed predominantly from states at the band edge, below those occupied by the nonthermal distribution excited by the pump. After 200 fs (dashed), the pump pulse is absorbed and

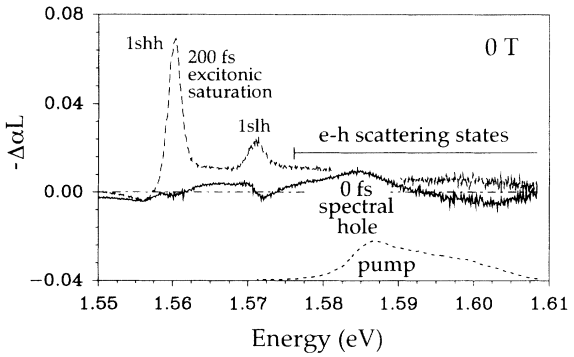

FIG. 2. Circular dichroism response, for $\Delta t = 0$ fs (solid) and $\Delta t = 200$ fs (dashed) for excitation 25 meV above the $1s$ hh exciton at zero field. Spectral hole in scattering states at $\Delta t = 0$ evolves into excitonic saturation at $\Delta t = 200$ fs as carriers thermalize. Spectral hole is burned at slightly lower energy than pump (dotted).

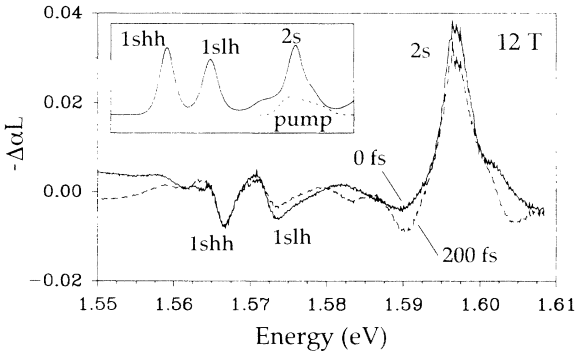

FIG. 3. Circular dichroism response, for $\Delta t = 0$ fs (solid) and $\Delta t = 200$ fs (dashed) for resonant excitation of $2s$ magnetoexciton at 12 T. Inset: Linear spectrum and the spectrum of pump (dotted), centered 25 meV above the $1s$ hh magnetoexciton. Saturation at $2s$ remains essentially unchanged as scattering out of state is suppressed.

thermalization complete, and the spectral hole has smoothed out to become a monotonically decreasing nonlinear signal in the scattering states. The PSF response of the excitons, however, rises very rapidly as the carriers thermalize to fill the states near the band edge. This signal, arising from the saturation of the excitonic resonance, results from the direct occupation of those scattering states, of small excess energy, required for the formation of the $1s$ excitons. The time-resolved CD signal therefore provides a direct observation of PSF effects during the thermalization of an initially nonthermal excitation. As with previous pump-probe transmission studies at zero field [9,10], the spectral hole-burning signal appears at an energy slightly lower than that of the pump photons (Fig. 2, dotted), and is accompanied by weak induced absorption at energies higher than the pump. These two effects arise from the Coulomb interactions between a probe *e-h* pair and the background photoexcited *e-h* plasma [10,11]. Induced absorption above the pump distribution results from the same Coulomb correlations that produce the Fermi-edge singularity observed in doped quantum wells [12]. The increased bleaching below the pump distribution results from a shift of the single-particle states to lower energy due to Coulomb attractions between electron and hole. The signals appearing at energies below the position of the spectral hole arise from PSF of the states composing the ground and excited states of the heavy- and light-hole excitons.

Confinement into 0D is effected by the application of a perpendicular magnetic field of 12 T. The absorption spectrum [3,13] consists of a series of sharp peaks separated by an energy of approximately 20 meV, indicating the 0D density of states for a material of excellent optical quality. An excitation centered 25 meV above the lowest magnetoexciton state is resonant with the first excited state of the magnetoexciton, the $2s$ state. By pumping resonantly with the $2s$ magnetoexciton (Fig. 3, inset), a nonthermal distribution is created by directly occupying this discrete state. The resulting CD response curves exhibit a strong PSF response at the $2s$, which is essentially unchanged on the time scale of the 2D thermalization. The signal at the $1s$ magnetoexcitons, a result of the phase space shared by the $1s$ and $2s$ states, is small and also unchanged on this time scale. Moreover, the magnitude of the screening response observed at the $1s$ magnetoexcitons, due to high field excitation of the $2s$, is much lower than that observed at the $1s$ in zero field under the same excitation conditions [3]. This occurs because the electrically neutral 0D magnetoexcitons provide only dielectric screening, while the 2D plasma screens very effectively by large amplitude motion of charge. Measurements on a longer time scale indicate that times on the order of 100 ps are required to scatter from the $2s$ to the $1s$ magnetoexciton at 12 T. Confinement into 0D inhibits the thermalization process, slowing by several orders of magnitude the redistribution of occupation for a 0D material excited out of thermal equilibrium.

The 25-meV excess energy of excitation for both 2D and 0D measurements is insufficient to allow the emission of transverse optical (TO) phonons [14]. Studies at zero field [4,15] indicate that the dominant mechanism for thermalization in intrinsic quantum wells excited with carrier densities at and above 10^{10} cm^{-2} is carrier-carrier scattering. This is consistent with the data presented here. Thermalization in excited 2D plasmas occurs through a very large number of carrier-carrier scattering interactions, each exchanging a small amount of momentum. Since Coulomb scattering favors small-angle interactions, the dense manifold of scattering states, each separated by very small energies, allows very rapid evolution of the initially nonthermal distribution. In 0D, however, all *e-h* pair states are bound by the effective potential associated with the magnetic field, so that the *e-h* scattering states are eliminated, and all optically active

excited states separated by energies of 20 meV [16]. Thermalization then occurs through the coupling of these energetically distant states, resulting in dramatically lowered thermalization rates in 0D.

Time-resolved circular dichroism measurements of nonthermal distributions in GaAs quantum wells separate completely, for the first time, the effects of screening and phase space filling. The thermalization of an excitation with 25-meV excess energy occurs in approximately 100 fs in 2D, without scattering between angular momentum states. During thermalization, the evolution from spectral hole burning to excitonic phase space filling is clearly exhibited in the CD response. The scattering of *e-h* pair states through the closely spaced states above ionization threshold in 2D is eliminated in the transition to 0D, resulting in an inhibition of the thermalization rate by several orders of magnitude. Continuous tuning of the separation of states, by control of the magnetic field, should allow measurement of the crossover from 2D to 0D behavior in the thermalization of excited carrier distributions.

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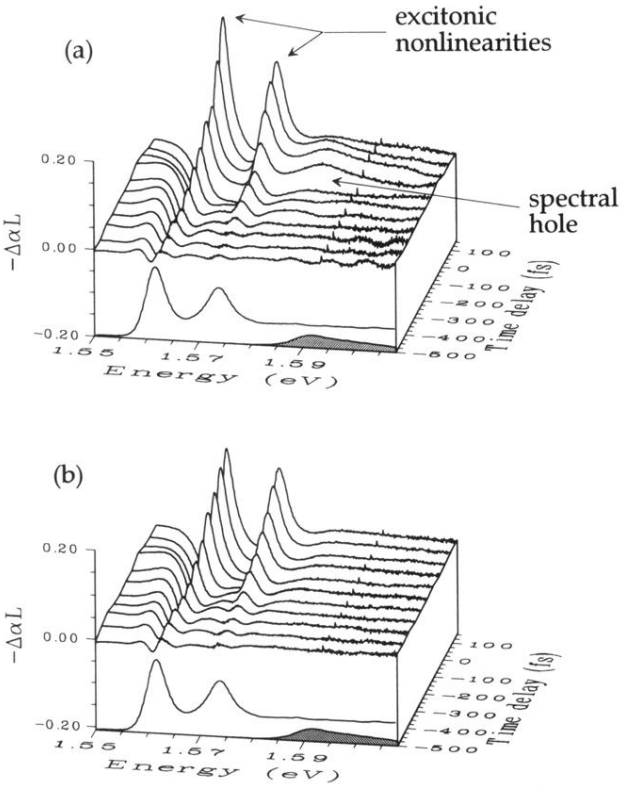

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