Room-Temperature Optical Nonlinearities in GaAs

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We report the first systematic study of the frequency dependence of optical nonlinearities of bulk GaAs at room temperature. In contrast to the previous understanding, band filling and plasma screening of Coulomb enhancement of continuum states are found to be the dominant contributions to the dispersive optical nonlinearities under quasi steady-state excitations. The partly phenomenological semiconductor plasma theory is in good agreement with the experimental data.

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There has been increasing interest in optical nonlinearities in semiconductors because of their underlying physics and potential applications to all-optical and electrooptical devices.[1] It is generally believed that the optical nonlinearity in InSb, for example, is mainly due to band filling,[2,3] whereas for GaAs the origin of the room-temperature optical nonlinearity responsible for optical bistability and optical logic gating has not been clearly understood, and has rather been attributed[4] to excitonic saturation.[5] At low temperature (2 K), excitonic contributions have been shown to be the dominant nonlinear mechanism.[5-8] As temperature increases, the exciton linewidth broadens because of the scattering with LO phonons.

Nonlinear-absorption changes at the exciton frequency were measured by Miller *et al.*[9] However, it is not clear if the nonlinear transmission change around the band edge is primarily of excitonic origin since the many-body nature of the plasma interaction has to be considered. To understand better the physics of the nonlinearity, accurate measurements of the nonlinear-absorption changes over a wide spectral range, including the band edge, are needed, as well as a proper semiconductor plasma theory. In this Letter we present the first systematic study of the frequency dependence of the optical nonlinearity in GaAs at room temperature. The microscopic origins of the nonlinearity were studied, and the maximum nonlinear dispersive changes ($\Delta n = -0.06$) in GaAs under quasi steady-state conditions were measured. In contrast to previous results, plasma screening of Coulomb enhancement of continuum states and band filling are found to be the dominant contributions to the dispersive nonlinearity. We have applied a partly phenomenological plasma theory (generalized Elliott formalism)[10] with excitation-dependent line broadening and find good agreement with the experimental data.

The experimental configuration was a pump-and-probe scheme utilizing an optical multichannel analyzer (OMA). The pump beam was tuned to 1.51 eV throughout the experiment and a broad-band (1.38–1.51 eV) probe source was obtained from photoluminescence of a 299-Å multiple-quantum-well (MQW) GaAs platelet pumped by an argon-laser beam. The pump and probe beams were synchronously modulated by acousto-optic modulators to produce 1-μs and 0.8-μs rectangular pulse trains, respectively, at 10 kHz. The transmitted probe pulses were collected and integrated by the OMA. Thermal effects up to the highest pump power (50 mW) were carefully checked and avoided by changes in the pulse widths and repetition rates. The temporal width of the pulse was much longer than the carrier lifetime, assuring quasisteady state for the duration of the pump. Spot diameters of the pump and probe beams were both 15 μm, but diffusion of the photogenerated carriers slightly increased the effective pump spot size to 20 μm. Small (< 3 μm) misalignments did not critically alter the absorption spectra. Bulk GaAs samples grown by molecular-beam epitaxy (MBE) consisted of a 2.05-μm-thick GaAs layer between two AlGaAs layers. For comparison purposes, 299-Å MQW GaAs (1.8-μm GaAs thickness) samples were also grown in the same chamber without our opening the MBE machine. Silicon monoxide antireflection coatings were deposited to minimize the Fabry-Perot effects from the surfaces of the samples.

The group of curves in Fig. 1(a) was obtained by variation of the pump intensity. The pump wavelength was set far from the band edge to minimize the effects of absorption saturation at the pump frequency. This non-

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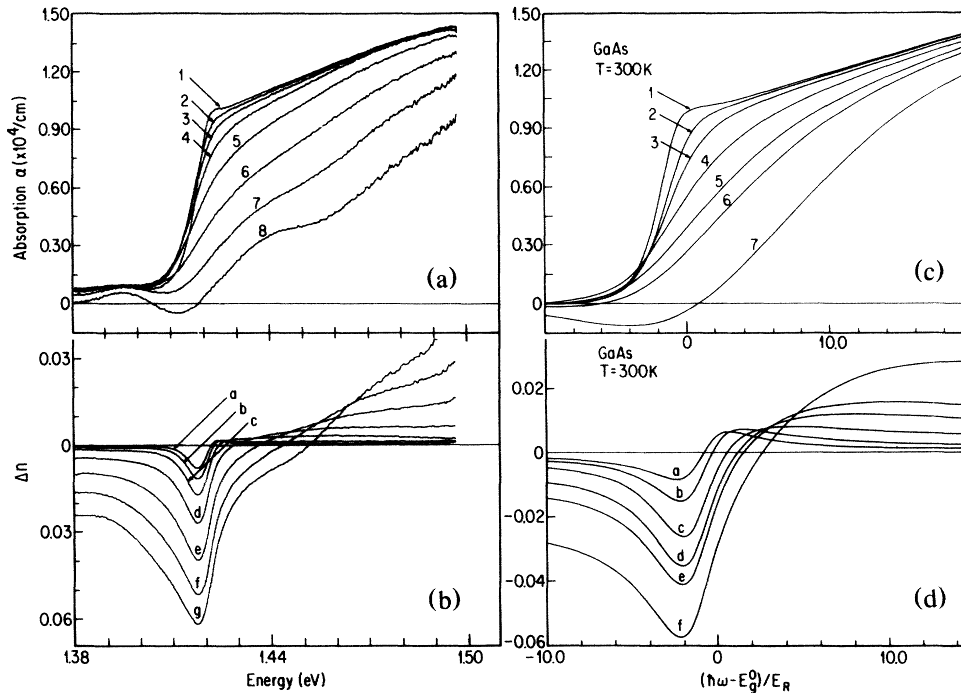

FIG. 1. Room-temperature bulk GaAs optical nonlinearities: experiment and theory. (a) Experimental absorption spectra for different excitation powers P: (1) 0, (2) 0.2, (3) 0.5, (4) 1.3, (5) 3.2, (6) 8, (7) 20, (8) 50 mW on a 15-μm diameter spot. (b) Nonlinear refractive index changes corresponding to the measured absorption spectra. Curves $a-g$ are obtained by the Kramers-Kronig transformation of the corresponding experimental data (2)-(8) in (a). (c) Calculated absorption spectra for different electron-hole pair densities N: (1) 10^{15}, (2) 8×10^{16}, (3) 2×10^{17}, (4) 5×10^{17}, (5) 8×10^{17}, (6) 10^{18}, (7) 1.5×10^{18} cm^{-3}. $E_g^0=1.420$ eV and $E_R=4.2$ meV. (d) Calculated nonlinear refractive index changes. Curves $a-f$ are obtained from curves (2)-(7) in (c), respectively.

resonant pumping generates an electron-hole plasma rather than excitons at room temperature. The top curve (1) in Fig. 1(a) is an unpumped absorption spectrum of bulk GaAs and shows a small exciton feature. At small pump intensities, most of the negative absorption change comes from the exciton saturation, but the net effect is not very strong. As the pump intensity increases, broad absorption changes in the band begin to take place and become dominant. The wavy structures in the absorption spectrum at highest pump intensity are due to the interference fringes arising from the incomplete antireflection coating (~1%). These structures are more pronounced at high pump intensities because the absorption is highly saturated. A Kramers-Kronig transformation of the measured absorption spectra yields a refractive index change $\Delta n(E)$, as in Fig. 1(b), given by

$$\Delta n(E)=\frac{\hbar c}{\pi}\mathrm{P}\int_{E_1}^{E_2}\frac{\Delta\alpha(E')}{E'^2-E^2}dE'.$$

The maximum refractive index change was approximately -0.06 at about 3-4 meV below the exciton energy. The rising slopes at the high-energy end of Fig. 1(b) are artifacts arising from the finite integration limits (1.38-1.50 eV) set in the Kramers-Kronig transformation.

The 299-Å MQW GaAs samples exhibited nonlinear behaviors similar to that of the bulk GaAs as shown in Fig. 2. Since the Bohr radius of the free exciton is comparable to the quantum-well thickness, the two-dimensional characteristics of MQW samples would be small, so that one would expect bulk GaAs behavior. The MQW sample showed well resolved exciton peaks up to the third subband. As in the bulk GaAs case, at low pump intensities, most absorption changes were from exciton saturation. At higher pump intensities, the saturation process was very similar to that of bulk GaAs. This relative similarity of nonlinear characteristics in bulk and 299-Å MQW samples explains the comparable hysteresis curves and switching powers for bistable operations in the two samples previously reported.[5]

Nonlinear-refractive-index changes were also directly measured by observation of the Fabry-Perot transmission-peak shift in a nonlinear etalon (finesse of 10) made of 299-Å MQW GaAs. To cross check this measurement we compare these data [circles in Fig. 2(b)] with the nonlinear index changes obtained by a Kramers-Kronig transformation of the absorption changes[11] under the same pumping conditions (1.51 eV). Below the band gap, nonlinear index changes of electronic origin showed blue shifts of the Fabry-Perot peak (fast

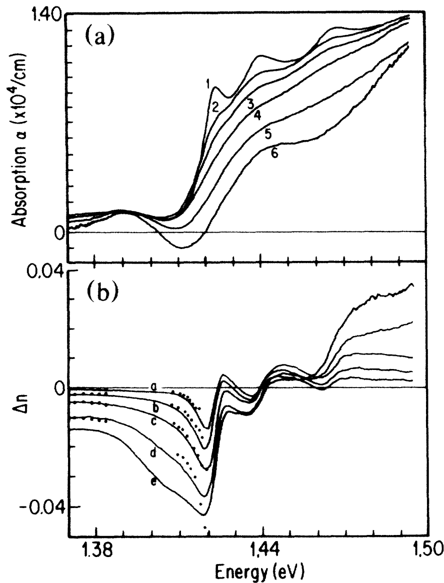

FIG. 2. Nonlinear absorption spectra of 299-Å MQW GaAs for excitation powers P: (1) 0, (2) 1.1, (3) 2.8, (4) 7.0, (5) 17.6, (6) 44 mW on a 15-μm diameter spot. Curves a–e in (b) are obtained from curves (2)–(6) in (a), respectively. Dots in (b) are the data obtained by measurement of the blue shifts of the Fabry-Perot transmission peaks.

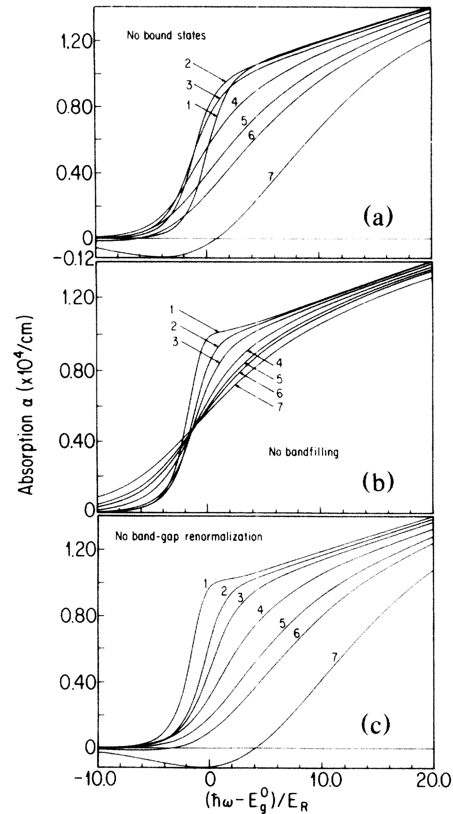

FIG. 3. The relative contribution of different nonlinear effects from the theory; labelling of curves as in Fig. 1(c).

negative index changes) and thermal index changes showed red shifts of the Fabry-Perot peak (slow positive index changes).

The experimental data are analyzed in the framework of a recently developed, partly phenomenological plasma theory in combination with excitation-dependent line broadening. This theory describes the semiconductor optical nonlinearities which are caused by a photogenerated electron-hole plasma. Because of the rapid interband relaxation times it is assumed that the electronic excitations have relaxed to quasiequilibrium distributions within their bands. At elevated temperatures, one can ascribe the density-dependent changes of the semiconductor properties to the effects of band filling, bandgap renormalization, and plasma screening of Coulomb interactions. Band filling reduces the valence-band to conduction-band transition probability because of the Pauli exclusion principle. Screening reduces the exciton binding energy (Mott transition), the Coulomb enhancement of the continuum states (Sommerfeld factor), and the semiconductor band gap. All these nonlinearities are incorporated in a generalized Elliott formula for the absorption,[10]

$$\alpha(\omega)=\alpha_0 A(\omega)\sum_\lambda |\phi_\lambda(\mathbf{r}=0)|^2 \times\delta_\Gamma(\hbar\omega-E_\lambda-E_g), \quad (1)$$

where α_0 is proportional to the absolute square of the interband transition matrix element, $A(\omega)=\tanh[(\hbar\omega-\mu)/2k_BT]$ is the band filling factor, $\mu=\mu_e+\mu_h$ is the sum of the quasichemical potentials of electrons and holes, $\phi(r)$ is the wave function for the relative motion of a (bound or unbound) electron-hole pair which is computed as an eigenfunction of the Wannier equation with the screened Coulomb potential, E_λ is the corresponding energy eigenvalue, and $E_g=E_g^0+\delta E_g$ is the density-dependent renormalized band-gap energy. The band-gap shift δE_g reproduces the Debye shift at low densities.[10] Also,

$$\delta_\Gamma(x)=E_R/\pi\Gamma\cosh(xE_R/\Gamma) \quad (2)$$

is the line-shape function which approximates the Urbach tail of the excitons, E_R is the exciton Rydberg energy, and $\Gamma=\Gamma_0+\Gamma_1 N$, where N is the electron-hole pair density. For GaAs at room-temperature, $\Gamma_0=1.25E_R$ and $\Gamma_1=(2\times10^{-18}\ \mathrm{cm}^3)E_R$ yields the results shown in Fig. 1(c). The corresponding dispersive changes (Δn) are obtained from a Kramers-Kronig transformation of the absorptive changes [Fig. 1(d)].

The relative simplicity of Eq. (1) allows one to analyze the relative contributions of the different nonlinear effects. Figure 3 shows absorption spectra obtained by artificially turning off (a) excitonic (bound state) contributions, (b) band filling, and (c) band-gap renormalization

during the calculation. The difference between Fig. 1(a) and Fig. 3 illustrates the relative contributions from the respective sources. Depending on the frequency regime, band filling and screening of the continuum-state Coulomb enhancement are the most efficient mechanisms responsible for the net dispersive changes: The band filling is responsible for the broad low-frequency tails of Δn, whereas the screening mainly causes the sharp structure in the vicinity of the band gap. The absorptive changes obtained through the bleaching of bound states (exciton ionization) are largely compensated through the red shift of the continuum states caused by the band-gap reduction. In other words, the negative Δn from exciton saturation is largely offset by a positive Δn from band-gap reduction at low intensities. The nonlinearities leading to a Δn sufficient for bistability and optical logic gating[12] are roughly equal contributions from the band filling and screening of the Coulomb enhancement of the continuum states.

In summary, we have analyzed the microscopic origins of the room-temperature optical nonlinearities in GaAs. In contrast to the previous understanding, the plasma screening of the continuum-state Coulomb enhancement and band filling are the dominant contributions to the dispersive nonlinearities responsible for optical bistability and optical logic gating in this material. The validity of the Kramers-Kronig transformation under quasi steady-state conditions was cross checked by two independent measurements of Δn and $\Delta \alpha$ under identical pumping conditions.

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