Femtosecond-Pump, Continuum-Probe Nonlinear Absorption in GaAs

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We present calculations of femtosecond-pump, continuum-probe nonlinear absorption in GaAs, including effects of electrons and heavy, light, and split-off holes. To account for hole-band nonparabolicity and anisotropy, a 30×30 $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian is diagonalized. Carrier dynamics are determined using an ensemble Monte Carlo method. Differential transmission spectra are obtained from the carrier distributions and directly compared with experiments. Our results show that pump–continuum-probe experiments provide the first direct evidence for a redistribution of holes on a femtosecond scale.

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Femtosecond laser spectroscopy experiments in GaAs have received much attention in recent years[1-6] because of their implications in the understanding of fundamental nonequilibrium carrier dynamics and because of their technological importance in the development of electro-optical devices. In particular, the fastest processes often cannot be measured by other means. Since important scattering times in GaAs are in the range of tens to hundreds of femtoseconds,[7] femtosecond optical experiments are well suited for studying ultrafast relaxation processes.

The recently developed femtosecond-pump, continuum-probe techniques of Schoenlein *et al.*[1] offer new flexibility by dramatically increasing the range of energy over which the carrier distributions can be investigated. However, interpreting the results from these experiments is difficult owing to the complexity of the carrier dynamics and optical transitions.

In this Letter, we present calculations of nonlinear absorption in femotosecond-pump, continuum-probe experiments[1] in GaAs. We calculate electron and hole dynamics by an ensemble Monte Carlo simulation and determine the nonlinear absorption using a $\mathbf{k}\cdot\mathbf{p}$ method. Our results show that pump–continuum-probe experiments provide the first direct evidence of a rapid redistribution of holes on a femtosecond scale, and also show that holes cannot be neglected in simulations of laser spectroscopy in semiconductors. Furthermore, we find that collisional broadening of the nonlinear absorption is an essential consideration in the calculation.

Our calculations also give a better understanding of ultrafast phenomena. For example, a rapid change in the transmission at 1.55 eV in the pump–continuum-probe experiments[1] was interpreted as evidence that a large population of electrons scatter within 200 fs to the bottom of the conduction band. However, other experiments, such as time-dependent photoluminescence in GaAs,[8] transient absorption in equal-energy femtosecond pump-probe experiments,[3] and subpicosecond Raman experiments,[9] indicate that it takes several picoseconds for most of the electrons to reach the band edge. Our results show that electrons relax slowly to the bottom of the band, but by including holes we obtain a quick change in the transmission in agreement with Schoenlein *et al.*[1]

Calculating the transient nonlinear absorption in GaAs involves several steps. Because of the large range of energies involved and because the heavy hole is significantly anisotropic even at low energies, the effective-mass approximation for the holes is inadequate. Therefore, we first calculate the hole band structures by diagonalizing a 30×30 $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian.[10] The matrix elements for the $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian are determined so that the band gap, spin-orbit splitting, and carrier effective masses agree with experimental values.[11,12] Thus, we take into account nonparabolicity and anisotropy of the hole bands, effects that are crucial for an accurate calculation of absorption away from the band edge. The $\mathbf{k}\cdot\mathbf{p}$ model is also used to determine the Bloch overlap factors and density of states for calculating hole scattering rates. In addition, it is used to determine the joint density of states $\rho_v(\hbar\omega)$ and matrix elements $H_v(\hbar\omega)$ for the optical transitions.

The initial carrier distributions are determined from the nonlinear absorption,[13]

$$\alpha(\hbar\omega,t)=\int d\hbar\omega\int dt' N(\hbar\omega,t-t')\sum_v |H_v(\hbar\omega)|^2\rho_v(\hbar\omega)[1-f_c^e(\hbar\omega,t')-f_v^h(\hbar\omega,t')]\,. \tag{1}$$

In this expression, the v and c subscripts denote valence (hole) and conduction bands, respectively, t is the delay between the pump and probe pulses, $N(\hbar\omega,t)$ is the photon density of the probe pulse as a function of energy and time, and $\sum_v$ represents a sum over the allowed transitions from the heavy, light, and split-off hole bands.

Collisional broadening is important and is included in the absorption by Lorentzian broadening with a spread of $\hbar/\tau$, where $1/\tau$ is the total scattering rate. Because the density of states for the holes is much larger than for the electrons, without collisional broadening the initial distribution peaks of the holes are much narrower than those of the electrons. As a consequence, the calculated initial transient of the differential transmission is unrealistically large. Collisional broadening spreads the initial distributions, which is especially important for the holes. Note that we include collisional broadening only in the calculations of the nonlinear absorption (pump and probe); i.e., collisional broadening effects are neglected in the carrier dynamics.

The evolution of the photoexcited carrier distributions is found by solving a set of coupled, time-dependent Boltzmann equations using an ensemble Monte Carlo method.[14,15] Scattering mechanisms treated include polar optical phonon, deformation potential, electron-electron, electron-hole, and hole-hole, with all possibilities of intra- and inter-hole-band scattering. Carrier-carrier scattering rates, which depend on the carrier distributions, are continuously updated. We use commonly accepted scattering-rate and band-structure parameters.[10,11,15,16] In addition, we assume that the laser pulse has a 20-meV Gaussian energy profile and a 75-fs hyperbolic secant-squared time dependence.

Results from the Monte Carlo simulation are shown in Fig. 1. The number of carriers per unit energy for the Γ-valley electrons and the heavy, light, and split-off holes are plotted as a function of carrier energy and time relative to the center of the 2.0-eV pump pulse. The electrons in Fig. 1(a) show three initial peaks, one for each of the transitions from the heavy, light, and split-off hole bands. The two peaks at high energy quickly diminish as electrons scatter into the L and X valleys. The lower-energy peak spreads owing to electron-electron and polar-optical-phonon scattering. At long times, a knee develops at 0.3 eV as a result of electrons returning from the bottom of the L valleys, and the density at the bottom of the band increases as electrons eventually thermalize. This is consistent with the experiments[3,8,9] that show, for high-energy photoexcitation, that most electrons relax to the band edge on a picosecond scale. The details of scattering to the satellite valleys and the effects of carrier-carrier scattering are discussed elsewhere.[15,17,18]

Although Fig. 1(a) shows the electrons slowly relaxing to the band edge, Figs. 1(b)–1(d) show the holes quickly redistribute to thermal-like distributions. Structure in the initial photoexcitation peaks in the light and split-off bands is mainly due to strong deformation-potential phonon scattering, which is not allowed in the Γ valley for the electrons.[7] The depletion of split-off and light holes at the bottom of the bands at long times is due to interband scattering, primarily to the heavy hole band.

After the carrier distributions are determined, the differential absorption, $\Delta\alpha = \alpha - \alpha_0$, is found from Eq. (1). α_0 is the linear absorption, i.e., the absorption with $f_c^e = f_v^h = 0$. If the width of the sample, W, is thin relative to the absorption length, then the differential transmission is related directly to the change in absorption by

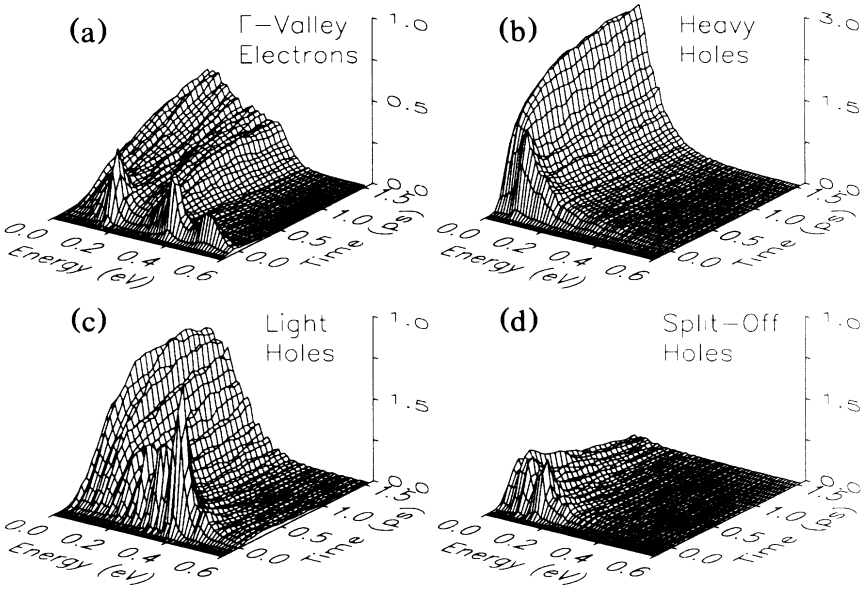

FIG. 1. Carrier density as a function of energy and time for (a) Γ-valley electrons, (b) heavy holes, (c) light holes, and (d) split-off for photoexcitation by a 75-fs pulse centered at $t=0$. Normalization for the z axis is arbitrary, but the relative normalizations between all figures are the same. In (a) the initial peaks at 0.16, 0.37, and 0.50 eV are from the three allowed transitions to the conduction band from the split-off, light, and heavy hole bands, respectively. Holes thermalize quickly, as shown by (b)–(d).

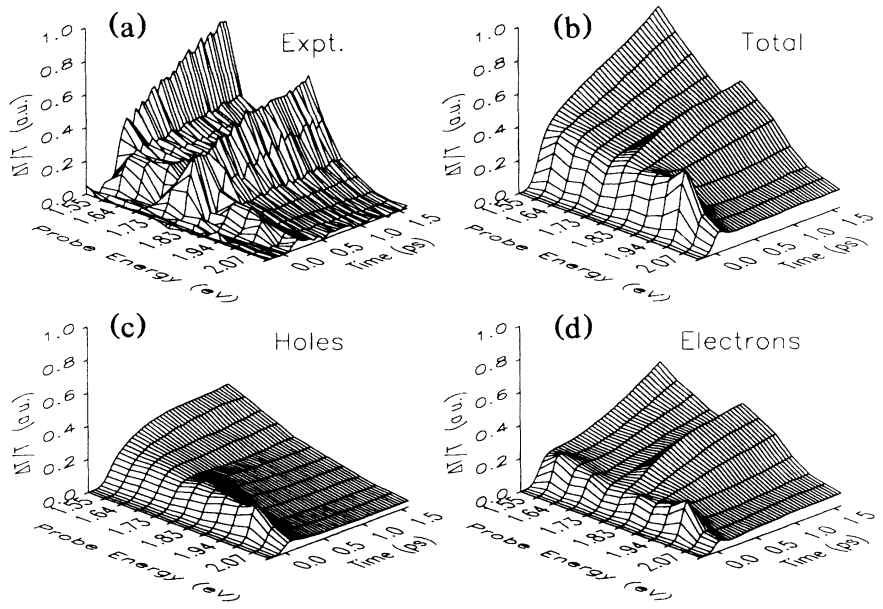

FIG. 2. (a) Experimental differential transmission spectra from Schoenlein *et al.* (Ref. 1) compared with (b) calculated differential transmission spectra including both electrons and holes, (c) including only holes, and (d) including only electrons. Neither (c) nor (d) alone match the experimental measurements as well as (b). Electron dynamics alone cannot completely explain the experiments.

$\Delta T/T_0 \approx \Delta\alpha W$.

Results for the differential transmission as a function of probe delay and energy are shown in Fig. 2 along with the experimental results of Schoenlein *et al.*[1] The pump energy is 2.0 eV, whereas the probe energy varies from 1.55 to 2.14 eV. Experimental data are shown in Fig. 2(a). Figure 2(b) shows the results of our Monte Carlo calculations including electrons and heavy, light, and split-off holes. Agreement between theory and experiment is qualitatively excellent and quantitatively good, especially since the scattering rates and band-structure parameters were *not* optimized to achieve the best fit.

Figures 2(c) and 2(d) show the contributions to the differential transmission of only the holes and of only the electrons, respectively. The hole differential transmission rises quickly with little overshoot before becoming flat. The electron contribution is different, showing a slow, increasing rate of rise in the contribution at low energy. The differential transmission for the electrons is lower at 1.73 eV than at 1.78 eV because the split-off transition is not allowed at the lower energy. These figures show the important and often overlooked role of holes in optical experiments. Neither the electron nor the hole response alone matches the experimental behavior. It is necessary to include both electrons *and* holes.

In Fig. 3, we show the individual contributions to the differential transmission at 1.55 eV. The nonlinear terms in Eq. (1), f_c^e and f_v^h, are separated into their constituent optical transitions. The solid and dashed lines give the differential transmission due to electrons in the conduction band probed by the heavy and light hole transitions, respectively, whereas the dash-dotted and dash-double-dotted lines are for holes in the heavy and light hole bands, respectively. There are no optical transitions from the split-off band for 1.55 eV in GaAs. The holes make a major contribution to the total differential transmission. Part of the initial rise in the solid line for

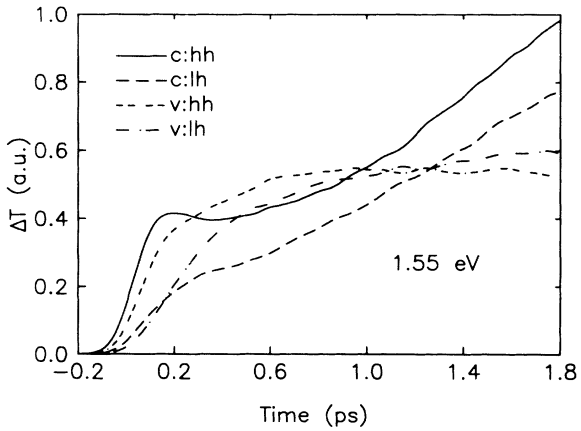

FIG. 3. Components of the differential transmission for a 1.55-eV probe pulse. Optical transitions probing the electrons are given by the solid line (heavy holes) and the dashed line (light holes). The components due to the presence of holes in the valence band are shown by the dash-dotted line (from the heavy hole) and the dash-double-dotted line (from the light hole).

the electrons occurs because the 1.55-eV heavy-hole transition probes part of the 2.0-eV split-off transition in the conduction band. Therefore, the initial rise in the differential transmission at low energy is due to a rapid redistribution of holes and because the 1.55-eV pulse probes the split-off transition of the pump pulse, *not* because of the rapid scattering of a large number of electrons to the bottom of the band.

In conclusion, we have presented the first calculations of the femtosecond-pump, continuum-probe nonlinear absorption in GaAs including the effects of holes, anisotropic and nonparabolic band structure, and collisional broadening in the optical transitions. Our results show that Schoenlein *et al.*[1] are the first to directly measure the rapid redistribution of holes on a femtosecond scale. We have shown that both holes and collisional broadening of the nonlinear absorption are essential and cannot be neglected. Although femtosecond nonlinear-absorption experiments are difficult to interpret, they can provide valuable insights into ultrafast relaxation processes when combined with detailed calculations of the electron and hole dynamics.

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