Subpicosecond Spectral Hole Burning Due to Nonthermalized Photoexcited Carriers in GaAs

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Subpicosecond infrared pulses were used to study in GaAs at 15 K the spectral dependence of the absorption saturation around the excitation-pulse wavelength. The nonthermal part of the carrier energy distribution is found to peak at the excitation frequency. A line-shape analysis allows the determination of the electron-hole-pair dephasing time.

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The dynamics of hot photoexcited carriers in semiconductors has been a subject of recent interest,[1-7] stimulated by the development of picosecond, and now femtosecond, lasers. The processes that govern the carrier dynamics are best investigated by first generating these particles in a nonequilibrium state with a short laser pulse, and then observing their subsequent relaxation. Most studies on the picosecond time scale have shown that carrier-carrier and carrier-phonon interactions lead to a thermalized (hot) energy distribution in a very short time scale, usually shorter than the pulse duration. With femtosecond lasers, ultrafast relaxations have been observed in various semiconductors. However, very little information has been obtained yet on the earlier steps of carrier relaxation, before the electrons and holes have been scattered out of their initial optically coupled states. We report here experiments performed with femtosecond pulses which give clear evident of the nonthermal electronic distribution and provide new information on the carrier dynamics.

Electron-hole (e-h) populations can be monitored through the band-to-band absorption saturation. Before evolving towards a quasiequilibrium, the e-h distribution is expected to be peaked at the excitation energy,[8] giving to rise to a dynamical spectral hole-burning effect, i.e., a preferential bleaching at the excitation frequency. Generally speaking, such a spectral hole burning yields information on the dephasing time and the cross relaxation of individual levels in a broad absorption band. This hole-burning effect has been observed for carriers in semiconductors only for intervalence-band transitions in p-type Ge crystal,[9-11] restricting the results to the case of holes. For electrons and holes, in most cases investigated up to now, the absorption saturation at photon energies slightly higher than the band gap primarily comes from the dynamical Burstein-Moss effect, i.e., *band filling* by a thermalized electron-hole plasma.[4] In our experiment, the interest has been centered on *state filling,* leading to the appearance of a dynamical spectral hole burning.

We report here the observation of such a hole burning in GaAs at 15 K, associated with the absorption saturation of valence- to conduction-band transitions by subpicosecond pulses at an optical frequency slightly above the band gap. These pulses had a spectral bandwidth close to the Fourier-transform limit, and increased the sample transmission by filling states with an initially quasimonoenergetic distribution. The transmission changes were probed by a weaker test pulse of considerably larger bandwidth with adjustable relative delays between excitation and test pulses. The excitation-pulse energy was tuned at 1.538 eV, i.e., only 19 meV above the band gap of the unexcited GaAs sample. This ensures that e-h pairs are created with an excess energy smaller than the optical-phonon emission, which leaves the intercarrier interactions as the dominant scattering mechanism.[7]

Our experimental setup is based on a laser system producing high-power tunable femtosecond pulses of 100-fsec duration.[12] The initial pulse at 620 nm is split into two parts, each focused into a 2-cm water cell to generate white-light continuum pulses. A narrow bandwidth is selected in one of the beams by an interferential filter, and further amplified in a styryl-9-dye solution, raising the available pump energy to the microjoule level. In the second beam, wavelengths shorter than 780 nm were filtered out. After transmission through the sample, this test beam was sent onto an optical spectrum analyzer made of a grating spectrometer followed by an SIT vidicon tube linked to an OSA 500 electronic processor for multichannel data acquisition. Comparison of transmitted versus incident spectra provided the normalized transmission spectra presented here on a logarithmic scale.

A key point in this experiment was the use of a narrow 5-meV-bandwidth (FWHM) pulse for the excitation, necessary for the clear observation of the spectral features reported here. Such a spectral narrowing resulted in some lengthening of the pump-pulse dura-

tion (0.5 psec), while the test pulse remained unchanged, allowing one to monitor the absorption changes during the excitation still with a 100-fsec resolution. Another experimental point of importance was the complete rejection of any light scattered from the pump beam by the sample surfaces. This was achieved by our setting the test-beam polarization perpendicular to that of the pump beam. An analyzer placed behind the sample could thus remove the major part of the scattered pump light. A further rejection was obtained by a careful spatial filtering with pinholes on the test-beam detection path.

The sample was a 0.75-μm-thick GaAs layer (residual doping 10^{15} cm^{-3}) sandwiched between two $Al_{0.3}Ga_{0.7}As$ layers grown by molecular-beam epitaxy on a GaAs substrate. A 1-mm^2 area of the substrate was selectively etched away, and antireflection coatings were deposited to avoid any Fabry-Perot interference fringes in the transmission spectra.

By variation of the relative delay between the test and pump pulses, the evolution of the transmission spectrum could be observed and was recorded with increments of 0.2 psec. A typical set of transmission spectra together with the pump intensity spectrum is shown in Fig. 1, at selected values of the delay and for a pump intensity of 100 MW/cm^2. The curve at 0 psec is characteristic of the unexcited sample (we arbitrarily took the zero time delay as just before the occurrence of the pump signal). Photocarriers induced by the test beam alone (50 kW/cm^2 · meV) cause some amount of exciton screening; as a result, the excitonic feature is broadened, but still apparent at 1.515 eV. Starting with the leading edge of the pump pulse, the sample transmission increases because of the absorption saturation, and a very clear feature centered at the pump wavelength is apparent, with a maximum amplitude at the top of the pump pulse. This feature disappears together with the excitation-pulse intensity, leaving in its place a smooth variation of the transmission as displayed in Fig. 1 for 4.2 psec. This suggests the following interpretation: (i) the occurrence during the pump pulse of a dynamical spectral hole burning, which corresponds to preferential state filling of the optically coupled states in the valence and the conduction band; (ii) the subsequent evolution towards band filling by a hot thermalized *e-h* distribution.

It is to be noted that a coherence coupling effect could have been observed if the pump and test beams were mutually coherent. However, these beams are obtained in two separate continuum cells, so that this

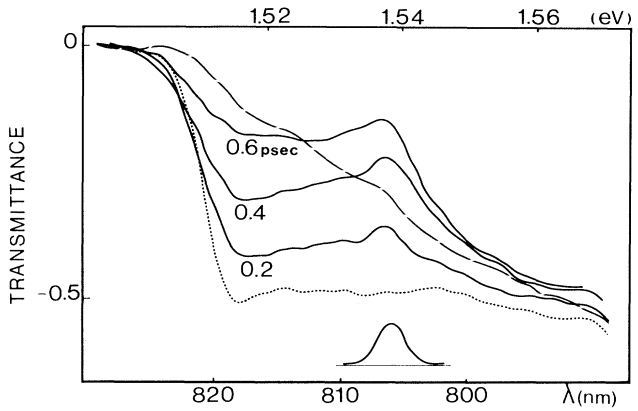

FIG. 1. Time-resolved transmittance spectra ($\log_{10}$ transmission) of GaAs at 15 K for different time delays between the pump and probe pulses. The dotted-line curve represents the transmission at $t = 0$ psec (just before the excitation). The dashed curve corresponds to a delay of 4.2 psec, a time significantly longer than the pump-pulse duration (0.5 psec FWHM). The pump-pulse spectrum is displayed in the lower part of the figure.

is not expected to be the case here. In addition, our use of orthogonal polarizations would also limit this contribution since no intensity interference can be produced by the superposition of the pump and test beams. We also note that the linewidth of the feature attributed to spectral hole burning is broader (3 times at peak intensity) than the pump linewidth. This shows that the dephasing time of the optically coupled *e-h* pairs can be deduced from our data, which we shall estimate with the following model.

Because of the momentum conservation accompanying the creation of each pair, the band-to-band transitions can be considered as an assembly of two-level systems with an energy separation $\hbar\omega$ and a density of states $\rho(\hbar\omega)$, also called the "joint density of states." In the absence of electronic collisions these two-level systems would be independent and behave like an inhomogeneously broadened absorption line. The frequency collisions experienced by electrons and holes lead to a very rapid cross relaxation between the two-level systems, leading to an internal thermal equilibrium. One can think of very sophisticated models to account for this cross relaxation, but these require quite complex numerical calculations.[8] Instead we propose a formulation which has the maximum simplicity but nevertheless accounts for the main features of the experimental observations.

The absorption coefficient at energy $\hbar\omega$ is expressed as

$$\alpha(t,\hbar\omega) \propto \int d(\hbar\omega')\rho(\hbar\omega')[1-f(t,\hbar\omega')]\frac{\Gamma}{(\omega'-\omega)^2+\Gamma^2}, \quad (1)$$

where Γ is the homogeneous linewidth of the individual transitions, and $f(t,\hbar\omega)$ is the sum $f_e + f_h$ of the electron and hole occupation numbers in the state optically coupled to the light at frequency ω. We now assume that $f(t,\hbar\omega)$ is the sum of a thermal distribution $f_{\rm th}(t,\hbar\omega)$, describing the *e-h* pairs which have already reached an

internal equilibrium, and a nonthermal distribution $f_{nt}(t,\hbar\omega)$, describing the e-h pairs that have just been photoexcited. The energy dependence of f_{th} is that of an e-h plasma,[4] with an initial temperature T_e corresponding to the equipartition between electrons and holes of the available excess energy $\hbar\omega - E_g'$, E_g' being the renormalized band gap. By contrast, the energy dependence of f_{nt} is linked to the optical field (it is peaked at the pump-photon energy). We identify $T_2 = 1/\Gamma$ as the dephasing time, consistent with the two-level framework already used for Eq. (1). This nonthermalized distribution also interacts with the thermalized system, and we assume for simplicity that this last contribution can be described by a lifetime T_1. This parameter T_1 is not the recombination time of the excited particles, which occurs on the subnanosecond time scale, but corresponds to the relaxation of the nonthermalized population towards the thermalized distribution. The rate equation for f_{nt} is then

$$\frac{d}{dt} f_{nt}(t,\hbar\omega) = \Omega^2\{1 - f(t,\hbar\omega)\}\frac{\Gamma}{(\omega-\omega_p)^2+\Gamma^2} - \frac{f_{nt}(t,\hbar\omega)}{T_1}, \tag{2}$$

where Ω is the Rabi frequency of the two-level transition, proportional to the amplitude of the optical field. In steady state, Eq. (2) can be solved for f_{nt}, leading to

$$f_{nt}(\hbar\omega) = \frac{\{1 - f_{th}(\hbar\omega)\}I}{1 + I + (\omega-\omega_p)^2/\Gamma^2}, \tag{3}$$

where $I = \Omega^2 T_1 T_2$ is the light intensity normalized to the saturation intensity $I_s = \pi\hbar^2\omega\rho(\omega)/\alpha_0 T_1 T_2$, with α_0 being the unexcited absorption coefficient. By inserting this result in Eq. (1) and assuming that the spectral variation of f_{th} is small over a homogeneous linewidth, one obtains the following expression for the absorption coefficient:

$$\alpha(\hbar\omega) = \alpha_0(\hbar\omega)\{1 - f_{th}(\hbar\omega)\}\left[1 - \left(1 - \frac{1}{(1+I)^{1/2}}\right)\left(\frac{1}{1+\Delta^2}\right)\right], \tag{4}$$

where

$$\Delta = (\omega-\omega_p)/\Gamma\{1 + (1+I)^{1/2}\}.$$

The last term in the last bracket of Eq. (4) corresponds to the hole burning. Because of this term, $\alpha(\hbar\omega)$ presents a characteristic dip centered at the pump frequency ω_p, in the form of a power-broadened Lorentzian. At $\Delta = 0$ the hole-burning contribution is equal to $1/(1+I)^{1/2}$, a well-known result for inhomogeneously broadened absorption lines.[13]

To interpret our data in terms of the this model, we have calculated theoretical curves of $\alpha(\hbar\omega)$ using Eq. (4), and adjusted the relevant parameters so as to obtain the best fit with the experimental curves. The comparison was restricted to photon energies higher than 1.52 eV, since at lower energies one should take into account the screening of the excitonic feature. The thermalized distribution[14] was calculated as in Minot *et al.*[15] For practical reasons, the comparison was done on the difference spectra $D(\hbar\omega) = (\alpha_0 - \alpha)/\alpha_0$, which should be equal to $f_{th}(\hbar\omega)$ in the absence of the hole-burning effect. A good fit was obtained for a single value of the dephasing time, i.e., $T_2 = 0.3$ psec (from other attempted fits, the accuracy of the T_2 determination is found to be within $\pm 30\%$) and a single value of electronic temperature $T_e = 120$ K. Figure 2 shows a comparison between the experimental and calculated values of $D(\hbar\omega)$ together with the values of N and I, which were treated as adjustable parameters for each time delay. One notices that though I and N have been treated as parameters, their variations are strictly determined: I follows the pump time profile, and N can be computed consequently. The values of I and N behave as expected.

The agreement between theory and experiment is satisfactory, in view of the simplicity of the model presented here. It can be seen from the data that the contribution of the thermalized population is important even during the leading edge of the pulse. This shows that T_1 is in fact quite smaller than the pulse duration, justifying the steady-state approximation used for Eq. (3). Additional calculations based on the measured depth of the spectral hole lead to an estimate for T_1 of 0.2–0.3 psec with use of Eq. (3) and with the assumption of negligible recombination on the T_1 time scale. The experimental data display a systematic small deviation from the calculated curve at energies immediately below the pump energy. This is probably the trace of the intermediate regime of carrier relaxation, in the evolution of the e-h distribution from the purely Lorentzian $f_{nt}(\hbar\omega)$ toward the thermal one $f_{th}(\hbar\omega)$, a regime not taken into account in our simple model.

Our determination of T_2 is in the range of the expected values, although close to the upper limit.[16] However, if T_2 were significantly smaller than our estimate, the hole burning would have been much broader. Since in our experimental conditions the thermalization should occur essentially through the e-e, e-h, and h-h collisions, one would expect the collision frequency to increase with the carrier density. This should appear as a gradual broadening of the hole-burning contribution as the carrier density increases during the pump pulse. Apart from the power broadening taken into account in our model, there is no such trend in the experimental data, which suggests a saturation of the collision frequency. This could ori-

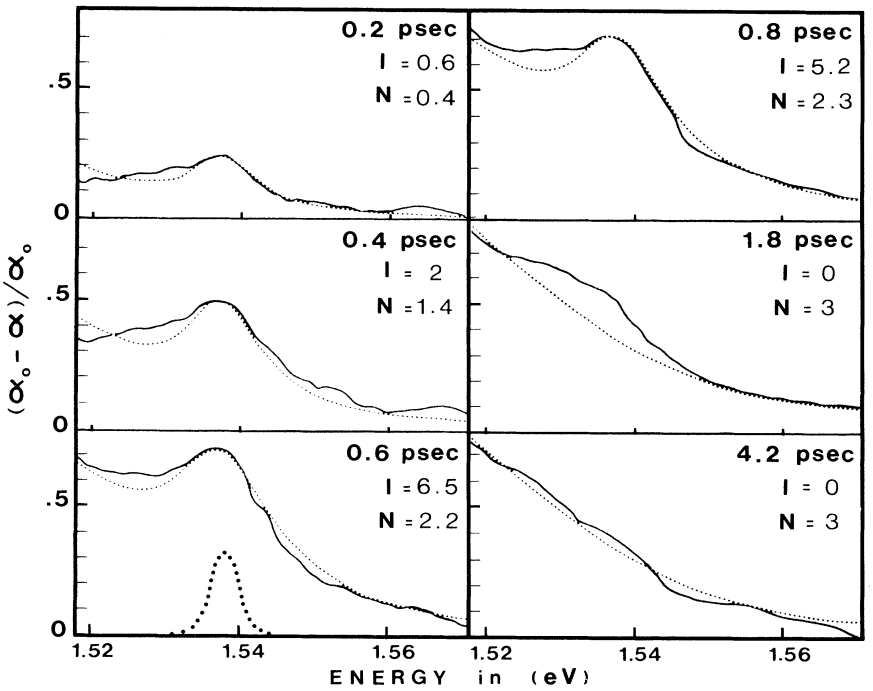

FIG. 2. Time-resolved difference spectra $[\alpha_0-\alpha(t)]/\alpha_0$, where α_0 is the unperturbed absorption coefficient, taken at different time delays t between the pump and probe pulses. The continuous lines are the experimental data, and the dotted lines are the best fits according to the model developed in the text. I is the pump intensity normalized to the saturation value, and N is the density of the thermalized e-h population in units of 10^{17} cm^{-3}. The heavy dotted line in the 0.6-psec data represents the pump profile.

ginate from the filling of the available density of states or from the screening of the Coulomb interaction. This screening is more and more effective with increasing carrier concentration, so that the reduction of the scattering cross section of individual carriers counterbalances the population increase. Finally, we note that subpicosecond pulses were necessary to observe such a clear hole-burning effect, since with longer pulses the relative magnitude of the nonthermalized contribution would be much smaller.

In conclusion, we have observed for the first time a spectral hole burning in GaAs in the valence-to conduction-band transitions. This was done with narrow-bandwidth subpicosecond pulses, which allowed the generation of an e-h plasma very far from equilibrium. The linewidth analysis made possible the determination of a dephasing time of 0.3 psec for the e-h pair transitions of low excess energy above the band gap.

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