Resonant Intervalley Scattering in GaAs

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The dynamics of intervalley scattering of electrons is investigated in GaAs with 6-fs optical pulses at room temperature. We report the observation of a "resonant coupling" between the Γ and X conduction bands. This effect slows the apparent rate of scattering of carriers in the Γ valley at energies near the minimum of the X valley.

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The process of intervalley scattering in semiconductors has been investigated extensively using optical techniques.[1-6] Femtosecond optical pulses have been used to investigate intervalley scattering from a highly nonequilibrium nonthermal population distribution in GaAs.[3-5] We report here the observation of the energy dependence of the $\Gamma \leftrightarrow X$ intervalley scattering time ($t_{\Gamma \leftrightarrow X}$) in GaAs using the broad spectral bandwidth of a 6-fs optical pulse. Previous hot luminescence measurements between nonthermal electrons and neutral acceptors have shown this energy dependence.[6] However, no direct temporal observation or detailed spectral dependence of this effect has been reported until now. Our measurements show that $t_{\Gamma \leftrightarrow X}$ is longer in a narrow energy range near the minimum of the X valley. These longer scattering times correspond to an apparent accumulation of carriers in the Γ band at an energy of about 30 meV below the X-band minimum. This effect, which is localized in a small energy range of about one phonon energy, will be termed *resonant intervalley scattering*. The experiment also reveals that during the first 50 fs after excitation by an ultrashort pulse, there is a fast transfer of carriers from the Γ to the X valley. This transfer which is much faster than LO-phonon-assisted scattering processes suggests that the electronic states are coupled in a different way during the early stage of the carrier redistribution created by an ultrashort laser pulse.

The experiment was performed at room temperature using the pump-probe configuration with 6-fs optical pulses. The optical pulses were obtained by compression of pulses produced by an amplified colliding-pulse mode-locked (CPM) laser[7] and have a maximum energy of 1 nJ centered at 620 nm. After pulse compression, the beam is split in a Michelson interferometer with an intensity ratio of 3 between the pump and probe beams, and focused into an 0.4-μm-thick free-standing GaAs sample which has been antireflection coated. The relative temporal delay between the pump and the probe beam is varied by a computer-controlled stepper motor. The induced change of transmission of the sample is recorded at different wavelengths by analyzing the probe spectrum in a monochromator. After the monochromator, the probe intensity is detected either with an optical multichannel analyzer or with a photomultiplier connected to a lock-in amplifier. The spectral bandwidth where the measurements have been performed is between 580 and 680 nm. The maximum density of photoexcited carriers is on the order of 10^{18} cm^{-3}. This density is deduced from the measured transmitted and incident beams, assuming that each absorbed photon excites one carrier in the conduction band. Measurements have been performed with linearly polarized beams, parallel to each other. In order to eliminate coherent coupling artifacts, measurements were also made using cross polarized beams. No significant difference in the results have been observed in these two configurations.

In Fig. 1 we have plotted the logarithm of the induced transmittance as a function of the time delay between the probe and the pump, for three different wavelengths. For each of these curves we have deconvolved the long transmittance recovery time (> 1 ps), which is due to the cooling of electrons to the lattice temperature.[8] In this figure, the decay is slower at 646 than at 636 and 656 nm, indicating a resonant behavior at $\lambda_{res} = 646$ nm. This wavelength corresponds to the creation of electrons with an excess energy of 0.45 eV in the Γ valley (for transitions from the heavy-hole valence band). The

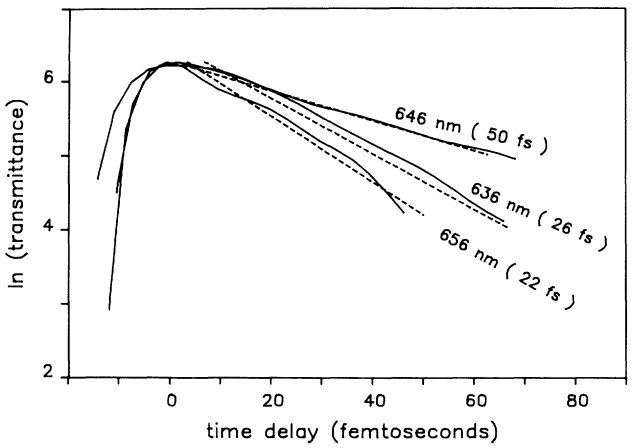

FIG. 1. Logarithm of induced transmittance as a function of probe delay for different wavelengths.

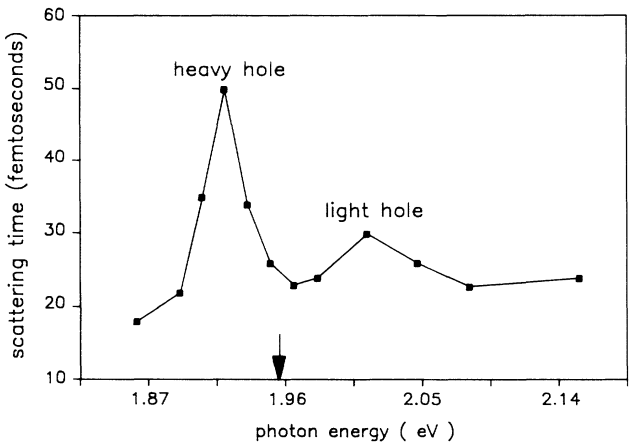

FIG. 2. Scattering time as a function of photon energies. The arrow shows the minimum of the X valley for optical transitions from the heavy-hole valence band.

same resonant slowing of the decay time at the wavelength λ_{res} was also observed with an attenuated pump beam which provided a total density of carriers of 4×10^{17} cm^{-3}.

A detailed spectral variation of the fast transmittance recovery time is shown in Fig. 2 where we have plotted the decay time derived as discussed for Fig. 1 as a function of the photon energy. This figure displays a variation of the recovery decay time from 20 to 50 fs over the entire spectral range. A clear peak occurs at 1.92 eV as well as an additional smaller peak around 2.015 eV.

In order to identify the two resonant peaks of Fig. 2, it is necessary to consider the different distributions of electrons by optical transitions from the heavy-hole (hh), light-hole (lh), and split-off (so) valence bands. With the broadband spectrum of the 6-fs pulse (580–680 nm), three distributions of electrons are created with excess energies in the Γ conduction band of 0.043–0.256, 0.287–0.566, and 0.358–0.637 eV for the transitions so$\rightarrow\Gamma$, lh$\rightarrow\Gamma$, and hh$\rightarrow\Gamma$, respectively. Since the minimum of energy of the X valley is 0.476 eV above the Γ minimum,[9] only the distributions created from the light- and heavy-hole bands cover the $\Gamma\leftrightarrow X$ transition point. We attribute the slower decay times at 1.92 eV (50 fs) and 2.015 eV (30 fs) in Fig. 2 to some additional population of electrons in the Γ valley which lies one LO-phonon energy below the X minimum. The two resonant peaks are due to the splitting of the two nondegenerate Γ_8 valence bands (heavy- and light-hole subbands). Their energy difference is about 95 meV. This is illustrated in Fig. 3 which shows optical transitions from the lh and hh bands which couple to the same energy in the Γ valley, 28 meV below Δ_X (i.e., one large-Q-vector LO-phonon energy below Δ_X). Thus, experimentally the resonant coupling between the Γ and X valleys is probed with two different wavelengths (λ_1 and λ_2 in Fig. 3). The energy splitting Δ_{hl} of 95 meV that we deduce from

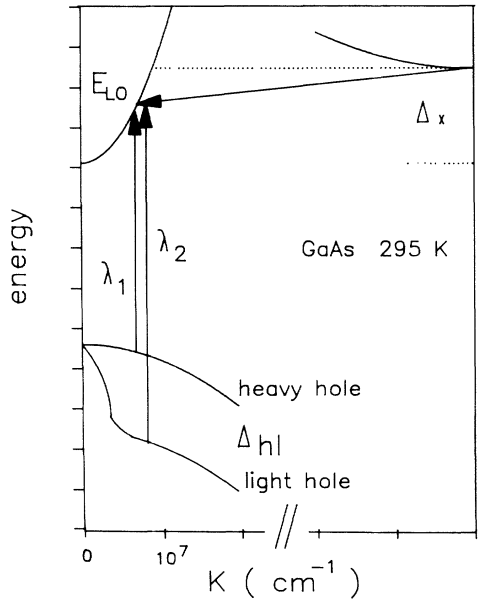

FIG. 3. Schematic of the optical transitions in GaAs indicating the spectral resonance. $\Delta_X=0.478$ eV is the difference of energy between the minima of the Γ and X valleys. Δ_{hl} is the splitting between the heavy- and light-hole valence bands.

Fig. 2 is consistent with the results of Ref. 6 where a strong energy variation of Δ_{hl} has been observed. Our technique, however, does not allow a fine resolution of the energy dependence of Δ_{hl} since we excite a wide energy range of carriers with the broadband femtosecond pulse. Therefore the 95 meV obtained here represents an averaged value of the splitting.

The apparent trapping of electrons in the Γ valley that we observe close to the X-valley minimum is a consequence of a resonance in the coupling of the two valleys. Our interpretation of this new effect is that because of the vanishing density of $|X\rangle$ states at the $\Gamma\leftrightarrow X$ transition point the return of electrons from the bottom of the X valley to the Γ valley is more probable, thereby enhancing the distribution of electrons in the Γ valley near the transition point. The experimental feature that supports this interpretation is the spectral variation of the intervalley scattering time $t_{\Gamma\rightarrow X}$ which is enhanced in a narrow bandwidth near the bottom of the X valley. In Fig. 2 there is a fast component in the spectral behavior of the decay time, which is of the order of 25 fs. Therefore a fast transfer of electrons to the X valley takes place immediately after excitation with the short pulse. This may be interpreted as a mixing of the Γ and X electronic states during this early stage of carrier redistribution.

In addition, the localization of the resonance about 30 meV below the minimum of the X valley suggests that the return of the electrons from the X valley in a LO-phonon-assisted process contributes to the resonant enhancement of the decay time. While most of the car-

riers in the X valley may thermalize through a succession of intervalley scatterings, those having an excess energy of less than one phonon with respect to the bottom of the X valley will more probably return to the Γ valley. Further evidence of this process is given by the width of the resonant peak which is on the order of one phonon energy. This interpretation of the apparent decay behavior that we measure seems to indicate that a round-trip of electrons from the Γ to the X valley and back to the Γ valley can be as fast as 50 fs.

From our measurements we estimate that, after 60 fs, less than 6% of the electrons remain in the Γ valley in the energy range probed in this experiment. Previous experiments[5,10] have shown that a part of the electrons also scatter to the L valley. The $\Gamma \leftrightarrow L$ transition point at 0.28 eV above the Γ-valley minimum[9] is outside the spectral region investigated and therefore does not influence the resonant feature observed in Fig. 2. However, $\Gamma \rightarrow L$ scattering processes may contribute to another resonant scattering process at an energy near the L-band minima outside our region of observation.

The different amplitudes of the two resonant peaks in Fig. 2 may also be understood by considering the different distributions of electrons that are probed. For photons with an energy of 2 eV, we probe the resonant $\Gamma \leftrightarrow X$ coupling from the light-hole valence band and also a fast component due to the electrons created from the heavy-hole band which scatter rapidly to the X and L valleys. For photons with energy 1.92 eV, we probe the resonance as well but we also probe a slower component due to the electrons created from the light-hole band and for which the scattering to the X valley is now forbidden.

In addition to the two resonant peaks in Fig. 2, one may notice the existence of a "background" with very fast scattering times on the order of 20 fs. Interestingly, this "background component" exhibits shorter times on the low-energy side (below the $\Gamma \leftrightarrow X$ resonance) than on the high-energy side. This feature is surprising since we may expect longer times below the resonant transition, where only $\Gamma \rightarrow L$ scattering channels are allowed, than on the high-energy side, where both $\Gamma \rightarrow L$ and $\Gamma \rightarrow X$ channels are open.

One may notice that the very short observed scattering times (20 fs at 1.87 eV) correspond energetically to a bandwidth of the order of 150 meV. This indicates that a broad spectral distribution of carriers is involved in this background component. We have no satisfactory explanation of this unexpected behavior at the present time. Experimentally, we could not probe lower energies to obtain more information below the resonance, without significantly distorting the temporal characteristics of our short pulse.

We would like to stress that when the experimental results of Fig. 2 are integrated with respect to the energy over the entire spectrum of the 60-fs pulse, one finds an average scattering time of 30 fs as measured previously.[5] Our results are also in good agreement with other measurements performed with a CPM laser operating at 620 nm,[3,4] where scattering times of the order of 30 fs were determined (see Fig. 2 at 2 eV).

We should also mention that our measurements as well as those reported in other experiments or calculations[2-5,10,11] differ significantly from results reported recently for carriers in equilibrium at low density.[6] Apart from the total density of created carriers which, as discussed earlier, does not affect the position of the resonant peaks, the main differences between our experiment and the one reported by Ulbrich, Kash, and Tsang[6] are the nonequilibrium distribution of carriers initially created with the ultrashort pulse, and the low temperature (2 K) at which their experiment is performed.[12] The resonant coupling reported here occurs during the first 60 fs after excitation and we believe that it could not be observed in an experiment involving a hot distribution provided by a cw laser beam. A good theory for this process does not exist. Such a theory should include the relaxation of electrons from a distribution corresponding to the pulse spectrum profile, to a hot Fermi distribution. Finally, we want to stress that, in agreement with previous observations,[1,6] we do observe a long return of the electrons from the X and L valleys (several picoseconds) for energies below the $\Gamma \leftrightarrow X$ and $\Gamma \leftrightarrow L$ transition points.

In conclusion, we have reported the existence of a resonant behavior in the spectral variation of the carrier relaxation dynamics in GaAs using 6-fs optical pulses. The resonance is characterized by a transient accumulation of electrons in the Γ valley at an energy below the X-band minimum. The resonance has been observed at two photon energies which correspond to the direct transitions from the heavy- and light-hole valence bands to the conduction band. We believe that this resonant trapping of electrons is related to the bottom of the X valley, where a fast return of electrons to the Γ valley at this particular energy is favored. This effect therefore reflects a resonant coupling between the two conduction bands at the $\Gamma \leftrightarrow X$ transition point.

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