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During the past year we have initiated a new research program to experimentally and theoretically investigate ultrafast processes and carrier dynamics in compound semiconductors. The objective of our program is to apply state of the art femtosecond measurement techniques including high resolution pump-probe absorption spectroscopy and time division interferometry measurements of nonlinear index as well as advanced theoretical techniques including ensemble Monte Carlo calculations and analytic solutions of rate equation models to study transient processes in semiconductors. The combination of experimental and theoretical approaches can be used to provide fundamental information about the physics of excited carriers in semiconductors and how they impact on electronic and optoelectronic device performance.

Experimental efforts at MIT have centered on the development of new techniques for measurement of ultrafast processes in semiconductors and their application to study transient processes in AlGaAs. Studies focus on both femtosecond measurements of nonlinear index as well as absorption. Investigations of nonlinear index are relevant for applications in all optical switching as well as high speed modulation using semiconductor devices. During the past contract year, we have developed a new femtosecond time division interferometry technique for characterizing nonlinear index changes from different components of the $\chi^{(3)}$ tensor. This new technique uses pump and probe measurements to investigate the amplitude and femtosecond dynamics of the nonlinear phase shift which is induced by a pump pulse on a probe pulse. Because nonlinear index effects are associated with below bandgap excitation, they are extremely small, and sensitive measurements using
waveguide geometries are required. We have developed a new time division multiplexed interferometer scheme which permits the measurement of nonlinear phase shift without parasitic effects from thermal index changes or acoustic vibrations. This new technique permits the characterization of the parallel and perpendicular components of the nonlinear index. Preliminary studies have been performed using optical fibers and semiconductor waveguides. Future investigations will apply this technique to characterize nonlinear index in compound semiconductors as well as quantum confined structures. Future studies will include investigations in the strained layer InGaAs/GaAs systems.

Other ongoing experimental work at MIT focuses on studies of femtosecond carrier dynamics in AlGaAs. We are currently performing pump probe measurements of carrier dynamics in AlGaAs using tunable 40 fs pulses from a high repetition rate femtosecond amplifier. Pump probe absorption saturation measurements are an indicator of the scattering carriers from their initial optically excited states. By varying the wavelength of the excitation pulses, the excess energy of the carrier distributions may be continuously varied. The objective of these studies is to investigate intervalley scattering by varying the carrier energies above and below the allowed energy for scattering from the \( \Gamma \) to the \( L \) valleys.

Finally we are beginning preliminary work on three pulse experiments to investigate the effect of a cold carrier distribution on the relaxation processes of hot carriers. In these measurements an initial electron and hole distribution is prepared by exciting the sample with a variable intensity ultrashort pulse. After a suitable delay to permit the excited characters to thermalize, a standard pump probe measurement is performed. Preliminary measurements have been performed using 2.0 eV pulses of 40 fs duration from a colliding pulse modelocked laser source. Future experiments using tunable pump and probe measurements will permit the investigation of both the scattering of excited carriers with a cold carrier background as well as the effect of high energy carrier excitation on a quasi-equilibrated electron in whole distribution. These experiments have direct bearing on gain and absorption dynamics in laser diodes where perturbations of quasithermal carrier processes produce changes in gain and index in high speed modulation.

Theoretical work at the University of Florida has focused on developing new approaches for studying ultrafast carrier dynamics of carriers photoexcited in GaAs and AlGaAs. Investigations have concentrated on Monte Carlo calculations including both the effects of electron and hole distributions and carrier-carrier scattering as well as analytic rate equation models of pump/continuum probe and tunable pump probe spectroscopy. Because the dynamics of excited carriers in semiconductors is extremely complex, detailed theoretical studies such as ensemble Monte Carlo or rate equation models are required to extract fundamental information on carrier dynamics from experimental measurements.
the past year, we have performed a number of theoretical investigations which are aimed at unifying experiment and theory.

We have developed ensemble Monte Carlo calculational techniques and correlated predicted measurements of pump and probe absorption saturation spectroscopy in GaAs and AlGaAs. These studies resulted in the identification and calculation of Coulomb enhancement effects from excitons near the band edge.

Other Monte Carlo studies have shown the importance of carrier diffusion effects in interpreting the role of carrier dynamics in both semiconductors. Theoretical investigations demonstrated that carrier diffusion can significantly effect the results of pump probe absorption saturation measurements of carrier-carrier scattering on the time scale of 10-100 fs. These investigations demonstrate that differences in sample thickness, even on the scale of 0.2-0.5 μm, affect the interpretation of experimental results by altering the carrier density profiles in the semiconductors.

Finally, we have developed an analytic solution of a rate equation model to describe intervalley scattering. The rate equation approach provides a simpler and computationally less complex model for interpreting femtosecond absorption saturation spectroscopic results. This model has a direct bearing on a number of previous experiments which attempt to measure intervalley scattering rates. Solutions of a rate equation approach show that the return time of electrons from the satellite L and X valleys is limited not by the intervalley scattering rate but is instead limited by inelastic scattering within the Γ valley (chiefly through polar optical phonon, POP emission). These results suggest that the accurate measurement of intervalley scattering must depend on the measurement of the initial scattering rate from Γ to L and not the return rate by which carriers return to the Γ valley.

Future theoretical studies will extend these results and develop a closer correspondence between ensemble Monte Carlo and rate equation models and femtosecond spectroscopic studies. The objective of these investigations is to extract fundamental information on carrier scattering events such as intervalley scattering and role of carrier-carrier scattering including electron hole distributions. Additional studies will extend these results to investigate coherent and non-Markovian processes, i.e., quantum corrections to the standard Boltzmann theory of carrier relaxation, at very short time scales of less than 15 ps.

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2. PUBLICATIONS


3. PAPERS IN PREPARATION


4. INVITED TALKS

1. Invited participant, Institute for Theoretical Physics Program on Mesoscopic Physics, Santa Barbara, CA, July 1991.

5. CONTRIBUTED TALKS (MEETINGS)


Femtosecond time division interferometry technique for measuring the tensor components of $\chi^{(3)}$

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We describe a new femtosecond time division interferometry technique for characterizing nonlinear index changes from different components of the $\chi^{(3)}$ tensor. Pump probe interferometric measurements of nonlinear phase are performed using a time division multiplexed reference pulse. The $n_{21}$ and $n_{23}$ components of the nonlinear index are measured by permuting the polarizations of the pulses and using a novel differential detection and modulation scheme. Measurements are demonstrated in an optical fiber.

Nonlinear index changes have been investigated using a number of experimental techniques including four-wave mixing, nonlinear waveguide couplers, nonlinear Fabry–Perot, fringe shift interferometry, Mach–Zehnder interferometry, and beam propagation distortion. In general, direct measurements of index nonlinearities are complicated because the measurement techniques are sensitive to thermal or acoustical parasitic signals, have low sensitivity, or require deconvolution.

Recently, we have developed a new technique, femtosecond time division interferometry (TDI), that permits high-sensitivity, direct measurements of the nonlinear index $n_2$ in waveguide devices. TDI was previously limited to measurements of $n_{21}$, i.e., index changes induced on a probe pulse by an orthogonally polarized pump. However, new materials such as organic polymers, multiple quantum wells and quantum wires are anisotropic and a complete characterization of $n_2$ requires the measurement of $n_{21}$.

Time-resolved measurements of $n_{21}$ are complicated by the fact that both the pump and probe pulses have the same frequency and polarization. In TDI measurements, this degeneracy prevents the interference signal between the probe and reference from being isolated from the interfering signals between the pump and probe or the pump and reference. In this letter we report a new extension of the time division interferometry technique that permits measurements of $n_{21}$. The contributions to $n_2$ from different components of $\chi^{(3)}$ can be characterized by measuring different permutations of pump and probe polarizations.

The laser source used for our experiments was a modelocked Nd:YAG which was pulse compressed and frequency doubled to synchronously pump a dye laser. Using the dye Styril 9, the system generates subpicosecond pulses tunable from 795 to 875 nm. The experimental configuration for time division interferometry has been described previously. A portion of the laser output is directed to a micrometer-stage delay line function as the pump. The pump beam is chopped and the phase delay of the pulse is modulated using a PZT attached to one of the delay mirrors. The probe and reference are obtained using a Michelson-type delay line with waveplates and a polarizing beam splitter. This system functions as a polarization sensitive delay line and generates orthogonally polarized probe and reference pulses. Half-wave plates placed in the probe, reference, and pump beams are used to set different combinations of polarizations for the three pulses. The pulses are then coupled into the waveguide structure to be characterized. After the waveguide, the signals are detected using a polarizer and dual detector. Finally, the phase bias operating point of the interferometer is actively stabilized using a PZT on the polarization-sensitive delay line. The feedback error signal is obtained using a boxcar integrator which detects the interferometer output signal when the pump pulse is blocked.

Figure 1 shows the relative positioning of the pulses for the measuring $n_{21}$ and $n_{23}$. The field amplitudes of the pump, probe, and reference pulses denote $E_{pu}$, $E_{pr}$, and $E_{ref}$. The pump pulse induces a nonlinear phase shift on the probe which is then measured by interfering the probe pulse with a time division multiplexed reference pulse. The reference pulse is then time delayed using a polarization-sensitive delay line so that it interferes with the probe pulse. For the measurement of $n_{21}$ the pump pulse does not have a different polarization and thus is not separated from the probe and reference pulses. The phase of the interferometer can be biased by adjusting the polarization-sensitive delay line with a PZT. The nonlinear index is characterized by measuring the phase shift of the probe relative to the reference using a polarizer and differential detector geometry. Finally, the dynamics of the nonlinear index may be measured by varying the delay between the pump and probe pulses.

The details of the measurement technique for $n_{21}$ and $n_{23}$ can be described by performing a simple analysis of the signals which are detected. Let us first consider the case where the pump and probe are perpendicularly polarized. The signals projected along the two polarization axes of the detectors are

$$S_{1,2} \sim |E_{pr}e^{i\phi_{nl}} \pm E_{ref}e^{i\phi_{bias}}|^2 + |E_{pu}|^2,$$

where $\phi_{nl}$ is the nonlinear phase shift produced by the pump on the probe, and $\phi_{bias}$ is the bias phase of the interferometer. In order to simplify the equations, we express the field amplitudes as real variables with relative phases expressed explicitly. The square of the field amplitude is...
normalized to intensity. Performing differential detection cancels the background intensity from probe and reference fields and yields the interference term:

$$ S = 4E_{pr}E_{ref} \cos(\phi_{nl} - \phi_{max}). $$

(2)

For measurements of nonlinear phase, the interferometer is biased to \( \phi_{max} = \pi/2 \), and the phase changes \( \phi_{nl} \) are small so the signal is given by

$$ S = 4E_{pr}E_{ref} \sin \phi_{nl} - 4E_{pr}E_{ref}\phi_{nl}. $$

(3)

Since the pump beam is chopped and \( \phi_{nl} \) is modulated, lock-in amplification detects only the terms that contain \( E_{pu} \). The contribution of these terms to the nonlinear phase shift is

$$ \phi_{nl} = \left( \frac{2\pi}{\lambda} \right) n_{21} E_{pu}^2 L_{eff} \left( 1 + \cos 2\phi \frac{\chi^{(3)}_{1221}(\omega;\omega, -\omega, \omega)}{2\chi^{(2)}_{1221}(\omega;\omega, -\omega, \omega)} \right), $$

(4)

where \( \phi \) is the phase between pump and probe and \( n_{21} \) is defined as the Kerr coefficient for monochromatic orthogonal polarizations with the coherence term suppressed. The term containing \( \cos 2\phi \) is the result of cross-phase modulation between the pump and reference. This term is averaged to zero when the pump phase is dithered using a PZT, or when the pump time delay is scanned. The resulting signal permits a measurement of \( n_{21} \).

$$ S = 4E_{pr}E_{ref}E_{pu}^2 (2\pi/\lambda) n_{21} L_{eff}. $$

(5)

Next let us consider the case where the pump and probe are polarized parallel to each other and interfere. For this case the detected signals in the two polarizations are

$$ S_{1,1} \sim |(E_{pr} + E_{pu} e^{i\phi}) e^{i\phi_{nl}} \pm E_{ref} e^{i\phi_{max}}|^2, $$

(6)

where \( \phi \) is the phase difference between probe and pump. Differential detection yields

$$ S \sim 8E_{pr}E_{ref}E_{pu}^2 (2\pi/\lambda) n_{22} L_{eff}. $$

(10)

Note that the detected signal for the parallel case has exactly the same form as in the perpendicular case except for an extra factor of 2.
In order to confirm that the measurement technique functions as predicted, experimental studies were performed using an optical fiber. The optical fiber is an isotropic material with well-established nonlinear optical properties. The sample consisted of 83 cm of Corning nonpolarization conserving fiber with a 5-μm core diameter. Special care was required in handling and mounting the fiber to avoid stress-induced birefringence effects.

In order to demonstrate the effect of pump phase, measurements were performed using parallel polarized pump and probe pulses, with and without dithering the pump phase φ (Fig. 2). Figure 3 shows the experimentally measured phase shift for the parallel and perpendicular pump and probe polarization configurations. The two measurements were performed by rotating the pump pulse polarization 90°. All other parameters, such as the probe and reference pulse polarization, the phase bias, φbias, and the intensities of all pulses were kept fixed. The pulse duration was 850 fs and the pump pulse energy was 5.5 pJ. Traces were averaged over several scans of the pump pulse delay with a data acquisition time of 2 min.

The ratio ν̂/ν̂ can be determined from the two curves. From (1) and (2), the ratio between the peaks of the two curves equals 2ν̂/ν̂. The measured ratio is 3.00 ± 0.07, which implies ν̂/ν̂ = 1.50 ± 0.04. This is in agreement with theoretical predictions based on symmetry considerations. For an isotropic medium, 11

\[
\chi^{(3)}(\omega, \omega, -\omega, \omega) = \chi^{(1)}(\omega, \omega, -\omega, \omega)
+ \chi^{(1)}(\omega, \omega, -\omega, \omega) + \chi^{(1)}(\omega, -\omega, -\omega, \omega).
\]

In silica fibers the dominant contribution to the nonlinear index is electronic and the three terms on the right-hand side of (11) are approximately equal. 12,13 The contribution from \(\chi^{(1)}\) yields a coherent interaction between the pump and probe pulses that averages to zero when the pump phase is dithered. Therefore,

\[
\frac{\nu\hat{\nu}}{\nu\hat{\nu}} = \frac{\chi^{(3)}(\omega, \omega, -\omega, \omega)}{2 \chi^{(1)}(\omega, \omega, -\omega, \omega)} = \frac{3}{2}
\]

The experiments agree with theory and yield the correct ratio for the two components of \(\nu\).

The experimental value for \(\nu\) can be obtained from the measured phase shift. In an experiment, the detected signal is averaged over the pulse shape and the transverse area of the waveguide. The measured peak of the detected signal is

\[
S = \frac{\int dt \int dA \, 8E_{pr}^2 \rho^2 E_{pu}^2 (2\pi\lambda) \nu_{\hat{\nu}} L_{eff}}{\int dt \int dA \, 4E_{pr}^2 E_{ref}^2 \rho^2}.
\]

The detected signal is normalized to the output of the interferometer when \(\phi_{bias} = 0\) and \(E_{pu} = 0\).

If the spatial field distribution in the fiber is approximated by a Gaussian, 14 and the temporal profile is also assumed to be Gaussian:

\[
S = \frac{2\pi (2\nu_{\hat{\nu}}) L_{eff} \sqrt{2ln2/\pi}}{\lambda \tau \omega^2},
\]

where \(\rho\) is the pulse energy, \(\tau\) the pulse FWHM, and \(\omega\) the field radius (3 μm for the fiber used). Using the results of Fig. 3, the measured value for \(\nu_{\hat{\nu}}\) is 3.31 ± 0.83 × 10^{-16} cm²/W. This is in close agreement with previous measurements of 3.2 × 10^{-16} cm²/W. 15 Finally, it should be noted that the measurement technique is extremely sensitive. The signal-to-noise ratio in Fig. 3 is greater than 10. Thus, phase shifts as small as 5 mrad can easily be detected. The authors would like to thank Kristin Rauschenbach for helpful scientific discussions. This work was supported in part by the Air Force Office of Scientific Research Contract No. F49620-88-C-0089, the Office of Naval Research Contract No. N00014-91-J-1956, the Joint Services Electronics Program Contract No. DAAL03-89-C-0001, and the National Science Foundation Presidential Young Investigator Award Grant No. ECS-85-52701.

Femtosecond temporal encoding in barium titanate

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We describe two-beam coupling and temporal encoding experiments in barium titanate. Volume gratings are created in the photorefractive material by 50-fs optical pulses. Information in the writing pulses may be encoded as spatially distributed volume gratings in the crystal. Femtosecond temporal waveform reconstruction is demonstrated.

The nonlinear-optical properties of photorefractive materials have been extensively studied in wave-mixing experiments, most of which have been conducted using cw lasers as light sources. Recently picosecond pulsed lasers with low and high repetition rates have also been used for the study of photorefractive materials as well as for coherence time measurements of the incident fields. The interaction of low-energy, high-repetition-rate pulsed lasers with photorefractive materials in the two-beam coupling configuration has been studied theoretically, and the resulting coupled equations have been solved in terms of the coherence functions of the incident fields. This theory describes multiple-pulse effects and the cumulative buildup of the internal electric fields that are responsible for the spatial modulation of the refractive index. It predicts that photorefractive properties are independent of the pulse duration, as long as the pulse energy and bandwidth are not excessive.

In this Letter, we describe the nonlinear interaction of femtosecond pulses with barium titanate using a dispersion-compensated colliding-pulse mode-locked (CPM) laser. We demonstrate femtosecond temporal information storage and explore the factors that determine temporal resolution. Experiments using stimulated photon echoes have demonstrated the possibility of storing temporal information in the frequency domain, with the use of inhomogeneously broadened atomic systems. The technique that we present here is more closely related to femtosecond holography. In femtosecond holography, a spatial distribution of interference gratings results from the different intersection points of the temporal features of the writing and reference beams. This is the approach taken by Dominic et al. to perform autocorrelation measurements of picosecond pulses using photorefractive materials. Conversely, it is also possible to use femtosecond holography to convert spatial features into temporal features.

We develop a simple geometrical model to describe our experimental results. The model involves parameters such as the beam waist at the crystal, the beam intersection angle, and the longitudinal length of the pulses (pulse duration multiplied by the speed of light). Similar models have been used to explain the results of an experiment with photorefractive materials in which beams having partial spatial coherence were used and can also be applied to describe temporal limitations of autocorrelation measurements of femtosecond pulses by noncollinear second-harmonic generation.

In order to explore the possibility of temporal information storage, we have used the experimental setup shown in Fig. 1. We use a reference beam and a signal beam. The CPM laser generates 50-fs pulses at a center wavelength of 625 nm and a pulse repetition rate of 100 MHz. A Michelson interferometer (M.I.) is used to generate a signal beam with pairs of copropagating pulses separated by a variable time delay. The average powers used in our measurements were typically 1 mW in the reference beam and 150 mW in the signal (double-pulse).

Fig. 1. Output of a CPM laser is prechirped by a grating pair and subsequently divided in two beams of variable relative delay. A boxcar averager, chopper, and autocorrelator (AC) are used to measure the duration of the pulses after the BaTiO₃ crystal.
blocked) experiment. The beam diameter at the crystal was set to ~2 mm. For $t_p = 50$ fs and $\theta = 3$ deg, $\omega_0/\cos \theta > l_p/\sin \theta$, and the mixing is in the large-beam-diameter case where the effective interaction length is smaller than the region of the beam overlap. Figure 3(a) shows the autocorrelation of the input pulses (after they pass through the crystal), while the autocorrelation of the scattered reconstructed pulse is shown in Fig. 3(b). There is a small but noticeable broadening in this case. On the other hand, when the two lenses ($f = 15$ cm) $L_1$ and $L_2$ are introduced, the beam waist is reduced to $\omega_0 = 30$ $\mu$m. Now $\omega_0 \cos \theta < l_p/\sin \theta$, and the mixing is in the small-beam-diameter case. In this case the autocorrelation of the scattered pulses shows almost no temporal broadening as seen in Fig. 3(c).

To demonstrate femtosecond temporal information storage, we use the double-pulse scheme de-
The incoming beams is greater than the crystal length (along the propagation direction).

In summary, we have performed the first studies of femtosecond induced gratings in a photorefractive material. Temporal information encoding and reconstruction have been demonstrated. Pulse broadening and temporal information storage behavior are commensurate with a simple geometrical model that uses spatially distributed volume gratings.

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References

Carrier diffusion effects in time-resolved photoluminescence

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We model time-resolved photoluminescence in GaAs using an ensemble Monte Carlo method coupled with a k-p calculation of the band structure. We show that on a picosecond scale, carrier diffusion perpendicular to the layer significantly reduces the density at the surface and consequently has a first-order effect on luminescence measurements. To illustrate this we compare the calculated luminescence, with and without diffusion, to experimental data.

Time-resolved photoluminescence, as recently demonstrated, is an excellent method for measuring carrier relaxation in GaAs. By measuring the time and energy dependence of the luminescence with subpicosecond resolution, electron and hole distribution functions can be probed on the time scale during which thermalization occurs. By studying these systems, detailed information can be obtained about band structure, scattering rates, and nonequilibrium relaxation processes.

Quantitative analysis of time-resolved luminescence in GaAs is difficult, however, owing to many different scattering mechanisms and complex band structure. In this paper, we show that an additional effect, carrier diffusion perpendicular to the layer, must be included to accurately interpret the luminescence spectra on a picosecond scale. That is, the density of carriers at the surface decreases in time due to diffusion into the sample. This has a striking effect on the shape of time-resolved luminescence.

Our results show that by omitting diffusion in the analysis, the carrier density is likely to be overestimated in time-resolved luminescence experiments. This can be seen from the evolution of the carrier spatial profile, and from the calculated distribution functions with and without diffusion. We show that diffusion has a strong influence on the luminescence at different energies and show that it must be included to match the experimental data.

Our calculational approach uses an ensemble Monte Carlo method to model photoexcited electron and hole dynamics. The hole band structure, optical matrix elements, density of states, region of photoexcitation, and Bloch overlap factors are determined from a 30-band k-p calculation. The initial photoexcited electron-hole pair states in the Monte Carlo simulation are determined according to the absorption of a monoenergetic pulse with a 0.5 ps full width at half maximum. The time-dependent distribution functions obtained from the Monte Carlo simulation are then used to calculate the time-dependent luminescence.

To include diffusion, the initial carrier spatial profile must be determined. The Monte Carlo method does not use a diffusion coefficient because the net redistribution of carriers depends directly on the transport parameters as given by the band structure and scattering rates. Thus, the main ingredient for diffusion in our calculations is an initial concentration gradient.

In time-resolved luminescence, the photoexcitation pulse is incident on the surface of a semiconductor sample. Lateral diffusion is unimportant in picosecond measurements if the spot size is chosen large enough. The spatial distribution of carriers perpendicular to the layer is given by the density gradient, where the density is lowest near the center of the pulse only half of the total number of carriers are excited. After completion of the pulse, the total average density in the layer is

\[ N_0 \frac{1-e^{-a_0 L}}{a_0 L} \]

where \( N_0 \) is the photoexcitation density, and where \( a_0 \), the linear absorption coefficient, is \( 4 \times 10^4 \text{cm}^{-1} \) for 2.04 eV photoexcitation in GaAs. That is, carriers are introduced into the Monte Carlo simulation according to the time dependence of the pulse at a k state and position determined by the absorption. For a pulse of finite duration, the carriers redistribute before the pulse is completed resulting in a more uniform profile.

The simulated spatial profile of photoexcited electrons is shown in Fig. 1 for a 0.5-ps pulse with \( N_0 = 3 \times 10^{18} \text{cm}^{-3} \) in a 4600-Å layer of GaAs. Transparent confining layers are assumed. The 0-ps curve in Fig. 1 is lowest because at the center of the pulse only half of the total number of carriers are excited. After completion of the pulse, the total average density in the layer is \( N_0 \frac{1-e^{-a_0 L}}{a_0 L} \), where \( L \) is the thickness of the layer. The initial distribution is nearly exponential because carriers have little time to redistribute during a 0.5-ps pulse. Within several picoseconds, however, the carriers diffuse away from the surface and reduce the maximum density by half.

As shown in Fig. 1, the surface carrier density is changing during the rise time because of diffusion. It first quickly increases with the excitation pulse, and then decreases as carriers redistribute. The changing concentration complicates analysis of time-resolved luminescence measurements.

The expression for the luminescence is obtained by summing the number of photons over allowed optical transitions,

\[ R(\omega, t) \propto \frac{1}{\omega} \int dt' W(t-t') \sum_{\nu} \int d\mathbf{k} |H_\nu|^2 \]

\[ \times \delta[E_\nu(\mathbf{k}) - E_\nu(\mathbf{k}) - \omega] f_\nu(\mathbf{k}, t') f_\nu(\mathbf{k}, t') \]  

(1a)
energies for a given f,(ot), of the pulse is set at 881
face of
then
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energy, and is not the same as the
nected region. A distribution functions averaged over the optically con-
ture length of 0.2
periments, however, the sample is more than twice the
nescence on a picosecond scale, in Fig. 2 we show
Since electrons provide most of the structure of lumi-
sorption length, then all carriers can contribute to the
surface. If
broadening'
in these simulations for
function with a full width half maximum of
ure the luminescence is sampled over a finite tempo-
measurement the luminescence is sampled over a finite temporal window given by W(t). A hyperbolic secant squared function with a full width half maximum of 0.5 ps is used in these simulations for W(t). We assume collision broadening
and band-gap renormalization
are unimportant in picosecond measurements and neglect these effects in our model. We also assume that only carriers at the surface contribute to the luminescence, since photons emitted deep in the sample are reabsorbed before reaching the surface. If the sample thickness is much less than the absorption length, then all carriers can contribute to the luminescence and there are no diffusion effects. In these experiments, however, the sample is more than twice the absorption length of 0.2 μm.

The terms of most interest in Eq. 1(b) are f,c(ω,t) and f,v(ω,t), the isoenergy distribution functions. These are the distribution functions averaged over the optically connected region. f(ω,t) is a function of the luminescence energy, and is not the same as the energy distribution function, f(E,t); if conduction and valence bands are isotropic, then f(E,t)dE = f(ω,t)do. The heavy-hole band for GaAs, however, is highly anisotropic. The isoenergy surface of ω is determined by the sum of electron and hole energies for a given k. A measure of the time-resolved luminescence, then, is a measure of carrier dynamics, albeit in a nontrivial manner.

In Eq. (1), the recombination of electrons and heavy holes dominates the luminescence. Heavy-, light-, and split-off hole dynamics are simulated, but the heavy holes control the luminescence because of their overwhelming number: at 1 ps after the center of the excitation pulse, the Monte Carlo simulation typically shows more than 95% of the hole population is in the heavy-hole band. Heavy holes have very high scattering rates, so convergence of the distribution function occurs nearly by the end of the laser excitation pulse, especially at high density where hole-hole scattering is strong. Electrons provide virtually all of the structure on a picosecond scale, however, because they cool much slower than heavy holes. Electrons are excited at higher energy than the heavy holes, and inelastic scattering rates are much lower. Electron relaxation is also slower because intervalley scattering prevents a direct descent to the bottom of the Γ valley.

Since electrons provide most of the structure of luminescence on a picosecond scale, in Fig. 2 we show f,c(ω,t) at the surface in the Γ valley for the two Monte Carlo simulations: (a) without diffusion, and (b) with diffusion. The area under f increases with time because the population in the Γ valley increases as carriers transfer from the satellite valleys. Comparing Fig. 2(a) with Fig. 2(b) shows that the occupation probability is less, and hence the surface density and the Fermi energy are less, when diffusion occurs. As Fig. 1 shows, the surface density is decreasing during the rise time of the luminescence. Diffusion thus has a significant influence on the distribution functions.

The importance of diffusion on the luminescence can
The data follow this pattern strictly only when diffusion is included. Curves are time shifted, but the time scale is preserved. Characteristics of the experimental data are an increasing rise time for decreasing energy. The data follow this pattern strictly only when diffusion is included.

Most easily be seen by comparisons at different energies. Figure 3 shows calculated luminescence for $3 \times 10^{18}$ cm$^{-3}$ photoexcitation in GaAs at 1.45, 1.57, 1.66, and 1.70 eV with and without diffusion, along with time-resolved luminescence measurements digitized from Ref. 1. Generally, rise time and decay time of the luminescence decrease with magnitude slower than the measurements. Any decay in the luminescence, therefore, is due to hot carriers relaxing and causing the luminescence to decay. At 1.45 eV there is no meaningful difference between luminescence rise times, because most states are occupied at low energy even with diffusion.

We have simulated time-resolved photoluminescence in GaAs by using a Monte Carlo method to model carrier transport and by calculating the recombination. Density of states, Bloch overlap factors, and optical matrix elements were determined by a k-p method to realistically include hole band-structure effects. Our results show that including carrier diffusion in the analysis causes the surface concentration to change on a picosecond scale and be significantly lower than without diffusion. We compared our calculated luminescence with experimental data and showed that diffusion is a critical consideration.

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**FIG. 3.** Calculated luminescence in GaAs at 1.45, 1.57, 1.66, and 1.70 eV for the first 20 ps after $3 \times 10^{18}$ cm$^{-3}$, 0.5-ns, 2.04-eV photoexcitation compared with measurements by Block et al. (see Ref. 1). Experimental curves are time shifted, but the time scale is preserved. Characteristics of the experimental data are an increasing rise time for decreasing energy. The data follow this pattern strictly only when diffusion is included.

With this in mind, Fig. 3 can be understood in terms of Figs. 1 and 2. Figure 1 shows diffusion decreases the surface density in the first 6 ps. At 1.70 and 1.66 eV in Fig. 3 this decreasing density accelerates the decay when diffusion is included. At 1.57 eV without diffusion, Fig. 2 shows that $f_s(\omega, t)$ is still increasing after 20 ps, thus causing the luminescence to steadily increase. With diffusion included, however, most of the states within a phonon energy below 1.57 eV are unoccupied, allowing electrons to continue relaxing and causing the luminescence to decay. At 1.45 eV there is no meaningful difference between luminescence rise times, because most states are occupied at low energy even with diffusion.

Rate equations for the study of femtosecond intervalley scattering in compound semiconductors

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We present solutions to a set of rate equations for the electron dynamics after photoexcitation by a 2.0-eV laser in GaAs and InP. Results obtained, although simpler than full Monte Carlo solutions, closely follow the experimental data and provide insight into intervalley scattering. Calculations show that the net return time of electrons from the satellite L valleys into the Γ valley is not limited by the intervalley scattering rate, but is instead limited by the polar-optic-phonon scattering rate within the Γ valley. This shows that the time-dependent mobility and luminescence experiments depend on the L-valley depopulation rate, which differs from the L → Γ intervalley scattering rate. Results further suggest that the Γ → L scattering rate is faster than the polar-optic-phonon scattering rate.

INTRODUCTION

Intervalley scattering in compound semiconductors is responsible for the transferred electron effect and is thus important in the operation of Gunn oscillators and microwave devices. As a result, there is interest in accurately measuring the intervalley scattering rates. With the development of ultrafast lasers, experiments with a time resolution comparable to the intervalley scattering times have become possible. These experiments include the rise time of band-edge photoluminescence, pump-infrared-probe absorption, femtosecond reflectivity, equal pulse correlation, pump-continuum-probe absorption, transient nonlinear absorption, and cw acceptor luminescence.

Several of these experiments claim to measure the deformation potential phonon scattering rate in GaAs for the scattering of carriers from the satellite L valleys back into the central Γ valley. The values reported for the deformation potential constant D_{rL}, however, have a large variance. In this paper, we show, through a series of rate-equation models, that the time measured in some of these experiments is not the L → Γ scattering rate, but the L-valley depopulation rate. This suggests that the apparent controversy is due in part to a misinterpretation of some of the experiments.

Comparison of our calculations to luminescence and mobility experiments shows that in GaAs, the L-valley depopulation rate is most strongly influenced by the rate of inelastic scattering, chiefly the polar-optic-phonon (POP) emission rate, within the Γ valley. If the inelastic scattering rate in the Γ valley is small compared to the Γ → L scattering rate, then to lowest order, the L-valley depopulation rate does not depend on the L → Γ rate. For this case, the bottleneck for the return of electrons from the satellite L valleys to the central Γ valley is the cooling of the electrons in the Γ valley. The cooling of electrons allows a net flow of electrons from the L valley into the Γ valley. In addition, our studies show that the Γ → L scattering rate is larger than the POP scattering rate.

This paper is organized as follows. We start with a two-state rate-equation model. While this model is too simple to describe optical experiments in GaAs, it illustrates an important point, namely that the two intervalley scattering rates Γ → L and L → Γ are not independent but are related through the density of states in each valley. We then extend our method to a three-state model. This allows us to describe actual experiments in GaAs provided that the measured quantities depend only on the valley the electrons occupy and not on the details of the electron states within each valley. (An example of such an experiment is the femtosecond mobility experiments of Nuss, Auston, and Capasso.) Finally, we extend the three-state model to a four-state model. With four states, we can account for experiments that are sensitive not only to which valley the electrons occupy, but also are sensitive to which state within the valley the electron occupies. For example, in the time-resolved luminescence experiments of Shah et al., only electrons at the bottom of the conduction band contribute to the luminescence signal.

TWO-STATE MODEL

We start with a two-state model shown in Fig. 1. It is the simplest possible model, but illustrates several key points about electron relaxation in compound semiconductors. One state represents all electrons in the Γ valley and the other represents all electrons that are in the L valleys. γ_{ΓL} is the Γ → L scattering rate, and γ_{LR} is the L → Γ scattering rate.

This is not a realistic model for carrier dynamics in compound semiconductors because it does not allow for states in the Γ valley that are not energetically able to scatter into the L valley. That is, relaxation of electrons...
in the \( \Gamma \) valley is ignored. Nonetheless, this model is instructive for the analysis of more complex systems and has features applicable to real systems.

The rate equations in this model are given in matrix form by

\[
\frac{d}{dt} \begin{bmatrix} n_\Gamma \\ n_L \end{bmatrix} = \begin{bmatrix} -\gamma_{\Gamma L} & \gamma_{\Gamma L} \\ \gamma_{\Gamma L} & -\gamma_{\Gamma L} \end{bmatrix} \begin{bmatrix} n_\Gamma \\ n_L \end{bmatrix}.
\]

(1)

Here \( n_\Gamma \) and \( n_L \) are the densities of electrons in the \( \Gamma \) and \( L \) valleys, respectively. A straightforward calculation shows that the eigenvalues and (unnormalized) eigenvectors are given by

\[
\lambda_1 = 0, \quad \lambda_2 = -(\gamma_{\Gamma L} + \gamma_{\Gamma L}),
\]

\[
V_1 = \begin{bmatrix} \gamma_{\Gamma L} \\ \gamma_{\Gamma L} \end{bmatrix}, \quad V_2 = \begin{bmatrix} 1 \\ -1 \end{bmatrix}.
\]

The zero eigenvalue results from the total density \( n_\Gamma + n_L \) being constant. The general solution to the two-state model is obtained by taking a superposition of the two eigensolutions:

\[
\begin{bmatrix} n_\Gamma(t) \\ n_L(t) \end{bmatrix} = A \begin{bmatrix} \gamma_{\Gamma L} \\ \gamma_{\Gamma L} \end{bmatrix} + B \begin{bmatrix} 1 \\ -1 \end{bmatrix} e^{-(\gamma_{\Gamma L} + \gamma_{\Gamma L})t}.
\]

(3)

The constants \( A \) and \( B \) are determined by the initial conditions:

\[
\begin{bmatrix} \gamma_{\Gamma L} \\ \gamma_{\Gamma L} \end{bmatrix} = \begin{bmatrix} n_\Gamma^0 \\ n_L^0 \end{bmatrix}.
\]

(4)

Here \( n_\Gamma^0 \) and \( n_L^0 \) are the initial populations of carriers in the two valleys. Solving Eq. (4) for \( A \) and \( B \), we obtain the solution

\[
n_\Gamma(t) = \frac{n_\Gamma^0 \gamma_{\Gamma L} - n_L^0 \gamma_{\Gamma L}}{\gamma_{\Gamma L} + \gamma_{\Gamma L}} e^{-(\gamma_{\Gamma L} + \gamma_{\Gamma L})t} + \frac{n_\Gamma^0 \gamma_{\Gamma L} - n_L^0 \gamma_{\Gamma L}}{\gamma_{\Gamma L} + \gamma_{\Gamma L}} e^{-(\gamma_{\Gamma L} + \gamma_{\Gamma L})t},
\]

(5)

\[
n_L(t) = \frac{n_L^0 \gamma_{\Gamma L} - n_\Gamma^0 \gamma_{\Gamma L}}{\gamma_{\Gamma L} + \gamma_{\Gamma L}} e^{-(\gamma_{\Gamma L} + \gamma_{\Gamma L})t} + \frac{n_L^0 \gamma_{\Gamma L} - n_\Gamma^0 \gamma_{\Gamma L}}{\gamma_{\Gamma L} + \gamma_{\Gamma L}} e^{-(\gamma_{\Gamma L} + \gamma_{\Gamma L})t}.
\]

The total number of carriers \( n = n_\Gamma(t) + n_L(t) = n_\Gamma^0 + n_L^0 \) is independent of time.

In equilibrium \((t \to \infty)\) the number of carriers in each valley is found to be

\[
n_\Gamma^e = n_\Gamma(t \to \infty) = \frac{n_\Gamma^0 \gamma_{\Gamma L}}{\gamma_{\Gamma L} + \gamma_{\Gamma L}} + \frac{n_L^0 \gamma_{\Gamma L}}{\gamma_{\Gamma L} + \gamma_{\Gamma L}},
\]

(6)

\[
n_L^e = n_L(t \to \infty) = \frac{n_\Gamma^0 \gamma_{\Gamma L}}{\gamma_{\Gamma L} + \gamma_{\Gamma L}} + \frac{n_L^0 \gamma_{\Gamma L}}{\gamma_{\Gamma L} + \gamma_{\Gamma L}}.
\]

From this, one obtains a "detailed balance" relation

\[
\frac{\gamma_{\Gamma L}}{\gamma_{\Gamma L}} = \frac{n_\Gamma^e}{n_L^e} = R
\]

(7)

\( R \) is defined to be the equilibrium ratio of the populations in the two valleys. If a more detailed model based on a complete description of all states were considered, then intervalley scattering events would obey a detailed balance relation given by

\[
W_{k'k}^{R} = \frac{W_{k'k}^{R}}{W_{k'k}^{R}},
\]

(8)

with \( W_{k'k}^{R} \) the transition probability per unit time from state \( k \) in the \( \Gamma \) valley to state \( k' \) in the \( L \) valley. This means that the \( \Gamma \to L \) and \( L \to \Gamma \) rates are dependent. Knowledge of one implies knowledge of the other.

Another point to note is that deviations from the equilibrium populations relax with a rate given by the sum of the two scattering rates \( \gamma_{\Gamma L} + \gamma_{\Gamma L} \). That is, if \( \delta n_\Gamma(t) = n_\Gamma(t) - n_\Gamma^e \), then

\[
\delta n_\Gamma(t) = \left( \frac{n_\Gamma^0 \gamma_{\Gamma L} - n_L^0 \gamma_{\Gamma L}}{\gamma_{\Gamma L} + \gamma_{\Gamma L}} \right) e^{-(\gamma_{\Gamma L} + \gamma_{\Gamma L})t}.
\]

(9)

This counters the naive assumption that a deviation from the equilibrium population in the \( \Gamma \) valley relaxes with \( \gamma_{\Gamma L} \) and a deviation in the \( L \) valley relaxes with \( \gamma_{\Gamma L} \). Any deviation from the equilibrium ratio of carriers will relax with the combined rate \( \gamma_{\Gamma L} + \gamma_{\Gamma L} \). While this seems strange for electrons in the \( L \) valley where the \( L \to \Gamma \) scattering rate is slower than the \( \Gamma \to L \) rate, it is important to remember that fewer electrons have to transfer from the \( L \) valley to restore the equilibrium ratio. Equation (9) means that the carrier densities in the two valleys equilibrate on a time scale comparable to the fastest of the two scattering times.

**THREE-STATE MODEL**

The two-state model is simplistic since it does not consider that the \( \Gamma \) valley is made up of several states. In particular, there is an energy threshold within the \( \Gamma \) valley below which electrons can no longer scatter into the satellite \( L \) valleys. A more realistic model for intervalley transfer must allow for this. Such a model is given by the three-state model shown in Fig. 2. In the three-state model, states in the \( \Gamma \) valley are separated above and below \( \Delta \), the energy threshold for transfer into the \( L \) valley. All \( \Gamma \) valley electrons with enough energy to transfer into the \( L \) valleys are in the \( \Gamma^\Delta \) state; all \( \Gamma \) valley electrons with energy less than \( \Delta \) are in the \( \Gamma^< \) state; and...
Rate Equations for the Study of Femtosecond...

\[
\Gamma \text{ Valley} \quad \xrightarrow{\gamma_{\bar{r}\Gamma}} \quad L \text{ Valley}
\]

\[
\gamma_\Delta \quad \gamma_{\bar{r}L} \quad \gamma_{r\bar{L}}
\]

FIG. 2. The three-state model for intervalley scattering. $\Gamma^>$ is the state that represents all electrons in the $\Gamma$ valley which are energetically able to transfer into the $L$ valley state. The $\Gamma^<$ state represents all electrons in the $\Gamma$ valley that do not have enough energy to transfer to the $L$ valley state. Scattering from the $\Gamma^>$ state to the $\Gamma^<$ state occurs via inelastic scattering within the $\Gamma$ valley with a rate $\gamma_\Delta$. This occurs mostly by POP emission.

Electrons in the $L$ valley are in the third state. Only electrons in state $\Gamma^>$ can scatter into the $L$-valley state and electrons in the $L$-valley state can only scatter into the $\Gamma^>$ state. Scattering from the $\Gamma^>$ state into the $\Gamma^<$ state is given by the scattering rate $\gamma_\Delta$ and occurs through inelastic scattering within the $\Gamma$ valley. This is chiefly through POP emission. While POP absorption makes scattering from below $\Delta$ to above $\Delta$ possible, for simplicity it is ignored in this model because POP emission occurs more frequently.

Note that for simplicity we have not included scattering into the satellite $X$ valleys. For 2.0-eV photoexcitation experiments, only a small fraction of the photoexcited electrons are energetically able to scatter into the $X$ valleys so that this is a minor correction. For higher energies, the $X$ valleys must be included, or the satellite valley state (and hence the appropriate scattering rates) modified to include both $L$ and $X$ valleys. Inclusion of an $X$ valley should be straightforward.

The equations for the three-state model are given by

\[
\frac{d}{dt} \begin{bmatrix} n_{\Gamma^>}(t) \\ n_L(t) \\ n_{\Gamma^<}(t) \end{bmatrix} = \begin{bmatrix} -(\gamma_{\bar{r}L} + \gamma_\Delta) & \gamma_{\bar{r}L} & 0 \\ \gamma_{\bar{r}L} & -\gamma_{\bar{r}L} & 0 \\ \gamma_\Delta & 0 & 0 \end{bmatrix} \begin{bmatrix} n_{\Gamma^>}(t) \\ n_L(t) \\ n_{\Gamma^<}(t) \end{bmatrix}.
\]

The eigenvalues are

\[
\lambda_0 = 0,
\]

\[
\lambda_\pm = -\frac{1}{2} \left( \gamma_{\bar{r}L} + \gamma_{\bar{r}L} + \gamma_\Delta \right) \pm \sqrt{(\gamma_{\bar{r}L} + \gamma_{\bar{r}L} + \gamma_\Delta)^2 - 4\gamma_{\bar{r}L}\gamma_\Delta}.
\]

The unnormalized eigenvectors for the matrix in the three state model are given by

\[
\begin{align*}
V_0 &= \begin{bmatrix} 0 \\ 0 \\ \gamma_\Delta \end{bmatrix}, \\
V_+ &= \begin{bmatrix} \lambda_+ \\ \gamma_{\bar{r}L}\lambda_+ \\ \gamma_{\bar{r}L}\lambda_+ + \lambda_+ \end{bmatrix}, \\
V_- &= \begin{bmatrix} \lambda_- \\ \gamma_{\bar{r}L}\lambda_- \\ \gamma_{\bar{r}L}\lambda_- + \lambda_- \end{bmatrix}.
\end{align*}
\]

The general solution is

\[
\begin{bmatrix} n_{\Gamma^>}(t) \\ n_L(t) \\ n_{\Gamma^<}(t) \end{bmatrix} = A n_0^{\Gamma^>} + B n_0^L + C n_0^{\Gamma^<} e^{\lambda_- t},
\]

with $A$, $B$, and $C$ determined by the initial conditions

\[
\begin{bmatrix} A \\ B \\ C \end{bmatrix} = \begin{bmatrix} n_{\Gamma^>}(0) \\ n_L(0) \\ n_{\Gamma^<}(0) \end{bmatrix}.
\]

Solving for $A$, $B$, and $C$ and using the relations

\[
\lambda_+ - \lambda_- = \gamma_{\bar{r}L}\gamma_\Delta,
\]

\[
(\gamma_{\bar{r}L} + \lambda_-)(\gamma_{\bar{r}L} + \lambda_+) = -\gamma_{\bar{r}L}\gamma_{\bar{r}L},
\]

we can find the final solution for the populations of the three states. For the experiments we are considering, electrons are initially excited only into the $\Gamma^>$ state, i.e., $n_{\Gamma^>}(0) \neq 0, n_L(0) = 0, n_{\Gamma^<}(0) = 0$; then

\[
n_{\Gamma^>}(t) = n_{\Gamma^>}(0) e^{\lambda_- t},
\]

\[
n_L(t) = n_{\Gamma^>}(0) \frac{\gamma_{\bar{r}L}}{\lambda_-} (e^{\lambda_- t} - e^{\lambda_+ t}),
\]

\[
n_{\Gamma^<}(t) = n_{\Gamma^<}(0) \frac{\gamma_{\bar{r}L}}{\lambda_-} \frac{\gamma_{\bar{r}L} + \lambda_-}{\lambda_-} (e^{\lambda_- t} - 1) - \frac{\gamma_{\bar{r}L} + \lambda_-}{\lambda_-} (e^{\lambda_+ t} - 1).
\]

Some insight into the intervalley problem is obtained by looking at the limiting form for the eigenvalues in Eq. (11). As mentioned earlier, the detailed balance relation requires that $\gamma_{\bar{r}L}$ and $\gamma_{\bar{r}L}$ be in the ratio of the equilibrium populations $R$. Taking into account nonparabolicity of the $\Gamma$ valley and the fourfold degeneracy of the $L$ valleys, for 2-eV photoexcitation in GaAs, electrons are photoexcited 0.5 eV above the bottom of the conduction band and
The scattering rate however, reflects the time dependence of with earlier Monte Carlo with time after the initial photoexcitation in agreement tion solution near ey, which is proportional to the total rates that can d ofagniude, hic as tome dclep uo electrons in the valleys, the two eigenvalues Furthermore, depending on the values for the scattering three-state model. The solid line is the experimental work of in spite of the fact that the scattering rates are constant. of a given state [given by Therefore the depopulation rate measure the L→Γ scattering rate. Only in this limit does the L-valley depopulation rate measure the L→Γ scattering rate.

It is interesting to note that, although the scattering rates given by the γ's in this model are constants, the densities given in Eq. (16) are not characterized by a single exponential decay. Therefore the depopulation rate of a given state [given by −d ln(n)/dt] is time dependent in spite of the fact that the scattering rates are constant. Furthermore, depending on the values for the scattering rates, the two eigenvalues λ- and λ+ can differ by more than an order of magnitude, which leads to depopulation rates that can change rapidly on a short time scale. Thus the total number of scattering events from the Γ to L valley, which is proportional to nΓ>(t)γΓL, changes rapidly with time after the initial photoexcitation in agreement with earlier Monte Carlo calculations. This change, however, reflects the time dependence of nΓ>(t) and not the scattering rate γΓL.

The three-state model can be applied to a large number of experiments that are sensitive primarily to which valley the electron populates. An example of such an experiment is that of Nuss, Auston, and Capasso (NAC). In the NAC experiment, the time-dependent reflectivity was measured on a femtosecond scale to infer the electron mobility as a function of time. Since the mobility of electrons in the Γ valley is nearly independent of energy, and the mobility of electrons in the L valleys is negligible by comparison, a measure of the mobility as a function of time is a measure of the density of electrons in the Γ valley (without regard to which state within the Γ valley the electrons occupy). In the three-state model, the mobility is therefore proportional to

\[
\mu(t) = \frac{nΓ>(t) + nΓ<(t)}{nΓ>}. 
\]  

To compare our calculations to the experiment, we must estimate the scattering rates. Based on the scattering rates of Schichijo and co-workers, we estimate the rates to be given by

\[
γΓL = 2 \times 10^{12} \text{s}^{-1}, \\
R \equiv \frac{γΓL}{nΓL} = 0.1, \\
γΔ = 5 \times 10^{12} \text{s}^{-1}. 
\]

In a later section, we show the sensitivity of the results to these values. The results of the three-state calculation are plotted in Figs. 3–6.

In Fig. 3, the experimental mobility from the NAC experiment is shown as the solid line. The calculated

FIG. 3. The mobility vs time for GaAs as calculated from the three-state model. The solid line is the experimental work of Nuss, Auston, and Capasso (Ref. 5), the dashed line is the result from the three-state model. It is assumed that the mobility of electrons in the L valleys is close to zero while the mobility of the electrons in the Γ valley is independent of energy. The mobility is therefore proportional to the number of electrons in the Γ valley [nΓ>(t) + nΓ<(t)]. The initial peak in the rate equation solution near t = 0 results from electrons initially photoexcited in the Γ valley that rapidly transfer into the L valley. This occurs on an extremely fast time scale and is not resolvable in the experimental data.
FIG. 4. Sensitivity of the rise time of the mobility to the inelastic (POP) scattering rate $\gamma_A$. The solid line is the experimental data (Ref. 5), the dashed line the results of the three-state rate-equation model. For the dash-dotted line, the rate is doubled, whereas for the dotted line, the rate is cut in half. As can be seen, the rise time of the mobility is very sensitive to this quantity.

(21)

To test the sensitivity of the rate equation model, we vary the parameters. In Fig. 4, we show the sensitivity of the rise time of the mobility to the POP scattering rate $\gamma_A$. The solid line corresponds to the experimental data of NAC, and the dashed line to the previous fit based on the rate-equation model (cf. Fig. 3). For the dash-dotted line, the POP rate is doubled, whereas for the dotted line, the POP rate is cut in half. As can be seen, the mobility and hence the number of electrons in the $\Gamma$ valley depends strongly on the POP scattering rate. If the POP scattering rate is low, $\gamma_A < \gamma_L$, then according to Eq. (19), it takes longer to get a net transfer of electrons back into the high mobility $\Gamma$ valley.

In Fig. 5, we show the sensitivity of the rise time of the mobility to the intervalley scattering rates. To do this, since we have shown that the $L \rightarrow \Gamma$ and $\Gamma \rightarrow L$ rates are dependent [cf. Eq. (7)], we vary both $\gamma_{TL}$ and $\gamma_{LT}$ but keep the ratio $R$ constant. The solid line corresponds to the experimental data and the dashed line the original fit. For the dash-dotted line, $\gamma_{TL}$ and $\gamma_{LT}$ are increased by a factor of 5, whereas for the dotted line, they are decreased by a factor of 5. As can be seen, the rise time is not strongly affected when the rates are increased, again consistent with Eq. (19), provided the ratio remains constant. The rate is divided by a factor of 5, however, then one is no longer in the limit $\gamma_{TL} \gg \gamma_A$ and a slight change in the time-dependent mobility is observed at short times. For slow intervalley scattering, i.e., $0.2 \times \gamma_{TL}$, there is not a rapid transfer of the initial electron populations into the $L$ valleys as in the other cases. If the $\Gamma \rightarrow L$ intervalley rate were this slow, one should see these effects in the experimental data. The fact that they are not seen shows that the $\Gamma \rightarrow L$ rate cannot be this slow and indicates that the intervalley rate $\gamma_{TL}$ is greater than the POP scattering rate $\gamma_A$.

In Fig. 6, we show the sensitivity of the rise time of the mobility to the density of states ratio $R$. The solid line is the experimental data and the dashed line is from the original calculated results. In all cases, we hold $\gamma_{TL}$ constant and vary $\gamma_{LT}$ to change the ratio $R$. For the dash-dotted line, the ratio is doubled, whereas for the dotted line it is halved. We can see that the data are very sensi-
ative to this ratio. In fact, the curves are similar to those in Fig. 4, which we should expect based on Eq. (19).

These results show that the $\Gamma \rightarrow L$ intervalley rate is faster than the POP rate. This agrees with the transient nonlinear absorption experiments of Rosker, Wise, and Tang,7 Schoenlein et al.,9 and Becker et al.,9,10 which predict fast $\Gamma \rightarrow L$ rates. Also, even though the $L \rightarrow \Gamma$ rate is slower than the POP rate, the return time of carriers from the $L$ valley is still limited by the relaxation of the electrons in the $\Gamma$ valley.

FOUR-STATE MODEL

The three-state model is useful for determining the $\Gamma$- and $L$-valley populations and is applicable for describing experiments which depend only on which valley the electrons occupy. Other experiments, such as the rise time of the band-edge photoluminescence, are more sensitive to the state within the $\Gamma$ valley that the electrons occupy. In particular, the photoluminescence experiments require that the electrons be at the bottom of the conduction band to contribute. Therefore the three-state model is not applicable. To account for this additional structure, we propose the four-state model shown schematically in Fig. 7. In the four-state model, an additional state is added in the $\Gamma$ valley representing electrons at the band edge and is labeled $\Gamma_{BE}$. The $\Gamma^-$ state now represents all the electrons with energies ranging from one optic phonon energy above the band edge to energies just below the threshold for intervalley transfer to the $L$ valley. For an electron to scatter to the top of the $\Gamma^-$ state to the bottom of the $\Gamma$ valley into the $\Gamma_{BE}$ state, requires that the electron emit approximately eight optical phonons (the first seven emissions keep the electron within the $\Gamma^-$ state). The scattering rate from $\Gamma^-$ to $\Gamma_{BE}$, denoted by $\gamma_{BE}$, is therefore approximately $\gamma_{\Delta}/8$.

In matrix form, the four-state model is given as

\[
\begin{pmatrix}
n_{\Gamma^+} \\
n_L \\
n_{\Gamma^-} \\
n_{\Gamma_{BE}}
\end{pmatrix}
= \begin{pmatrix}
-(\gamma_{\Gamma L} + \gamma_{\Delta}) & \gamma_{LL} & 0 & 0 \\
\gamma_{\Gamma L} & -\gamma_{LL} & 0 & 0 \\
\gamma_{\Delta} & 0 & -\gamma_{BE} & 0 \\
0 & 0 & \gamma_{BE} & 0
\end{pmatrix}
\begin{pmatrix}
n_{\Gamma^+} \\
n_L \\
n_{\Gamma^-} \\
n_{\Gamma_{BE}}
\end{pmatrix}
\]

From these equations, one obtains the solutions
FIG. 8. Rise time for the luminescence in GaAs and InP as calculated from the four-state model. The solid lines are from the experimental work of Shah et al. (Ref. 2). The dashed lines are the results of the four-state model. For InP, the intervalley scattering rates $\gamma_{IL}$ and $\gamma_{IL'}$ are set to zero keeping all other rates the same. As can be seen, the rate-equation model accurately predicts the observed differences between GaAs and InP. Differences between the experimental curves and rate-equation model at short times originate from the finite temporal width of the laser pulse not accounted for in the four-state model.

Results for the four-state model are shown in Figs. 8–12. The experimental luminescence is proportional to the density of electrons at the bottom of the band, i.e., the population of $\Gamma_{BE}$. In Fig. 8, we show the experimental data for the rise time of the luminescence in GaAs and InP from Shah et al. (solid lines). We also plot the results of the four-state model (dashed lines). We use a $\gamma_{BE}$ rate of $6 \times 10^{11}$ s$^{-1}$, approximately $1/2$ of $\gamma_\Delta$. For InP, since the $L$ valleys lie too high in energy for intervalley transfer in the Shah et al. experiment, we set the intervalley rates to zero keeping all other rates the same. As can be seen from the curves, the rate-equation model accurately predicts the dependence of the rise time of the luminescence for both InP and GaAs. Since the total number of electrons in the $\Gamma$ valley as a function of time does not change between the three-state and four-state models (only the occupation of the different $\Gamma$ valley states changes), the rise time of the mobility is exactly the same as before.

In Fig. 9, we check the sensitivity of the luminescence rise time to the POP scattering rate. The solid line is from the experiment, the dashed is from the original fit, and the dash-dotted and dotted correspond to doubling and halving the POP rate, respectively. Just as for the

Sensitivity to $\gamma_1$ GaAs

FIG. 9. Sensitivity of the rise time of the luminescence for GaAs in the four-state model to the electron-phonon scattering rate $\gamma_1$. The solid line is the experimental data of Shah et al. (Ref. 2) and the dashed line is the result of the four-state model. For the dash-dotted line, the electron-phonon scattering rate is doubled, while for the dotted line, the rate is halved. As can be seen, the rise time of the luminescence is very sensitive to this rate.

Sensitivity to $\gamma_1$ GaAs

FIG. 10. Sensitivity of the rise time of the luminescence in GaAs to the intervalley scattering rate. The solid line is the experimental curve of Shah et al. (Ref. 2), and the dashed line the result of the four-state model. For the dash-dotted and dotted lines, the intervalley scattering rates are changed so that the ratio $R$ remains constant. For the dash-dotted line the rate is increased by a factor of 5, while for the dotted line, the rate is divided by a factor of 5. As can be seen from the figures, the rise time of the luminescence is insensitive to the intervalley rates provided the ratio of the equilibrium populations is constant.

Sensitivity to $R$ GaAs

FIG. 11. Sensitivity of the rise time of the luminescence in GaAs to the density of state ratio $R$. The solid line is the experimental work of Shah et al. (Ref. 2) and the dashed line the solution to the four-state model. For the dash-dotted line, the ratio of the populations is doubled, while for the dotted line, it is cut in half. The curves are similar to those in Fig. 9, showing that the rise time of the luminescence depends on the rate $\gamma_\Delta$ (as well as $\gamma_{BE}$).
mobility, we see that the rise time of the luminescence is very sensitive to this rate. The net transfer of carriers from the \( L \) to the \( \Gamma \) valley depends strongly on the POP scattering rate within the \( \Gamma \) valley.

In Fig. 10, we show the sensitivity of the luminescence to the intervalley scattering rates keeping the ratio \( R \) constant. As before, there is not a strong dependence on the magnitude of the rate, even when divided by a factor of 5, provided the ratio is kept constant. We thus see that experiments measuring the rise time of the luminescence are insensitive to the intervalley rates, and only depend on the density of states in the two valleys and the inelastic scattering rate within the \( \Gamma \) valley.

In Fig. 11, we show the sensitivity to the ratio \( R \). Again, the luminescence is sensitive to this ratio as predicted by Eq. (19).

**CONCLUSIONS**

We have provided and solved a set of rate equations for intervalley scattering in compound semiconductors such as GaAs or InP. Using standard values for the transport parameters, solutions to these rate-equation models predict the experimental trends in time-dependent mobility and luminescence experiments quite well. Although these rate equations are simpler than full Monte Carlo modeling\(^7,8\) or numerical solutions to the Boltzmann equation,\(^25\) they nonetheless illustrate several key points.

(i) The \( \Gamma \rightarrow L \) and the \( L \rightarrow \Gamma \) intervalley scattering rates are dependent and related, through detailed balance, to the densities of states in each valley.

(ii) In time-dependent mobility and time-resolved photoluminescence experiments, one measures the depopulation rate of the \( L \) valley. The depopulation rate of the \( L \) valleys is not the same as the \( L \rightarrow \Gamma \) intervalley scattering rate unless the inelastic-scattering rate in the \( \Gamma \) valley is significantly larger than the \( \Gamma \rightarrow L \) rate. From an estimate of the transport parameters that are applicable to GaAs and InP, we find excellent agreement between the rate equations and experimental data for both the time-dependent mobility and the rise time of the luminescence. The scattering rates suggest that the \( L \)-valley depopulation is dominated by the energy relaxation of the electrons in the \( \Gamma \) valley. That is, POP scattering in the \( \Gamma \) valley acts as the bottleneck for the return of the electrons from the \( L \) valley. This return is not limited by the \( L \rightarrow \Gamma \) intervalley scattering rate. This suggests that time dependent mobility and band-edge luminescence experiments are not optimum for determining the intervalley scattering rates.

(iii) The initial low mobility in the NAC experiment can only be explained by a rapid transfer of electrons from their photoexcited states into the \( L \) valleys, suggesting a fast \( \Gamma \rightarrow L \) rate. This is consistent with the insensitivity of the rise time of the luminescence in GaAs to the \( \Gamma \rightarrow L \) rate, as well as the transient nonlinear absorption experiments of Rosker, Wise, and Tang,\(^9\) Schooelen et al.,\(^10\) and Becker et al.,\(^5,10\) which predict fast \( \Gamma \rightarrow L \) rates. Thus this is further evidence that the \( \Gamma \rightarrow L \) intervalley scattering rate is faster than the POP scattering rate.

While the rate equation models are simple, they nonetheless provide valuable insight into the qualitative carrier dynamics in compound semiconductors and isolate characteristics often lost in more detailed calculations.

**ACKNOWLEDGMENTS**

We are thankful to Frank Wise and Pradeep Kumar for useful discussions during this work. We are also grateful for the hospitality of the Institute for Theoretical Physics where part of this work was completed. This work was supported in part by the National Science Foundation through Grants Nos. DMR8957382 and PHY89-04035 and by the U.S. Office of Naval Research through Grant No. N00091-J-1956.

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RATE EQUATIONS FOR THE STUDY OF FEMTOSECOND ... 837


2.  
Studies of Intervalley Scattering using Tunable Femtosecond Pulses

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Abstract

We report experimental and theoretical investigations of intervalley scattering in Al-GaAs. Femtosecond absorption saturation measurements are performed using tunable 50 fs pulses and results interpreted using an ensemble Monte Carlo simulation as well as a rate equation model.
Studies of Intervalley Scattering using Tunable Femtosecond Pulses

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Summary

Intervalley scattering in AlGaAs has been studied by a variety of techniques. Subpicosecond and CW luminescence spectroscopy[1,2] and femtosecond absorption saturation[3,4] measurements have been used in attempts to determine the intervalley scattering rates and deformation potentials. While these measurements were the first to set limits on the intervalley scattering rates, they do not directly measure the intervalley scattering from the initial optically excited states and discrepancies exist between reported values. Most of these investigations measure the return of carriers from the satellite valleys by probing the occupancy of the Γ valley at energies near the conduction band edge. These measurements are strongly affected by inelastic scattering within the Γ valley[5].

We report investigations of intervalley scattering in AlGaAs using tunable femtosecond pulses. By systematically varying the wavelength and spectral content of the pulses, different scattering channels may be isolated, such as scattering to the L satellite valley.
Pump probe absorption saturation measurement permits a direct investigation of scattering from the initial optically prepared carrier distributions and allows a sensitive and direct measurement of intervalley scattering processes.

The laser source in our experiments is a dispersion compensated colliding pulse modelocked ring dye laser (CPM). 40 fs pulses from the CPM are amplified in a high repetition rate copper vapor laser pumped dye amplifier. The amplified pulses are focused on a jet of ethylene glycol to generate a femtosecond white light continuum. The continuum is filtered in the frequency domain with a grating Fourier filter[6] shown schematically in figure 1. This filter allows continuous control of both the center wavelength and bandwidth or duration of the femtosecond pulses. In addition, arbitrary temporal pulse trains may be generated by appropriate masking in the Fourier plane. Femtosecond transient absorption saturation is measured using pump-probe techniques with differential detection. Experiments are performed using antireflection coated 0.2 micron thick MBE samples of Al$_{2}$Ga$_{1-z}$As which are clad by Al$_{0.6}$Ga$_{0.4}$As.

The mole fraction of Al can be explicitly chosen to vary the band structure and select desired transitions. For example, in Al$_{0.1}$Ga$_{0.9}$As, scattering to the $L$ valley is energetically allowed for carriers photoexcited at energies of $\sim 1.8$ eV or greater (see Fig. 2). The use of frequency synthesis techniques for these measurements represents a powerful approach since the wavelength, pulse duration and the initial spectral distribution of carriers can be systematically controlled. This permits a comprehensive investigation of carrier dynamics. For example, figure 3 shows pump probe measurements of transient absorption saturation behavior in Al$_{0.1}$Ga$_{0.9}$As at 1.86 eV and at 1.72 eV using 50 fs pulses. The presence or absence of the intervalley scattering channel to the $L$ valley is manifest as a change in the behavior of the initial femtosecond transient in the data. Systematic measurements may be performed as a function of both photon energy and wavelength.

The determination of fundamental scattering parameters directly from pump probe data is complicated by the fact that several scattering processes and transitions are involved. Thus we use two theoretical approaches to interpret our data. The first technique
is based on a discretization of the Boltzmann transport equation and the scattering states which yields a simple rate equation model. This describes the carrier populations in the $L$ valley, high in the $\Gamma$ valley, and at the band edge. This model represents a compact and simple calculational approach which qualitatively predicts the effects of different carrier-carrier and intervalley scattering rates on the experimental results. However, in order to obtain a more comprehensive understanding of the carrier dynamics we have developed a full ensemble Monte Carlo simulation of the carrier dynamics. 40,000 electrons and holes are simulated and scattering mechanisms included are electron - polar optic phonon, electron - electron with time dependent screening, electron - intervalley, hole - non-polar optical phonon, and hole - hole. The distribution functions generated by the Monte Carlo simulation are used to predict differential transmission curves which may be compared directly to the experimental data. Monte Carlo techniques represent a rigorous approach for studying carrier dynamics since different experimental parameters can be used as inputs for the simulations. In addition, the effects of different scattering processes may be studied independently by isolating them in the simulation.

We gratefully acknowledge C.A. Wang from M.I.T. Lincoln Laboratory for preparing the MBE samples used in this investigation. *LHA's permanent address is Dept. de Fisica, Univ. Fed. de Pernambuco, Brazil.

References


Figure Captions

Figure 1. Schematic diagram of the experimental apparatus. A grating Fourier filter is used to control wavelength and bandwidth.

Figure 2. Schematic bandstructure of Al$_{0.1}$Ga$_{0.9}$As showing optical transitions above and below the L valley.

Figure 3. Transient absorption saturation data for pump-probe energies above (1.86 eV) and below (1.72 eV) the L satellite valley. The photoexcited carrier density is $\sim 10^{18}$ cm$^{-3}$. 
Figure 1
Figure 2
$\Delta T/T$ (%)

1.86 eV

1.72 eV

DELAY (PS)

Figure 3
Abstract Submitted
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Sorting Category 15B

Theory of Optical Gain in Silicon Quantum Wire Lasers. G. D. SANDERS, C. J. STANTON, University of Florida;* Y. C. CHANG, University of Illinois at Urbana-Champaign — We present theoretical calculations of gain, refractive index change, and differential gain for lasers made from regular arrays of silicon wires. We use a second-nearest-neighbor empirical tight-binding model to calculate subband energies and optical matrix elements. Laser properties are studied in a density matrix formalism with intrasubband relaxation. For narrow quantum wires, the silicon wire band gap is direct and the oscillator strength for interband transitions are comparable to that of direct gap materials such as bulk GaAs. Thus the enhanced optical matrix elements combined with an enhanced density of states leads to large optical gain. We find laser performance is sensitive to wire size and polarization.

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Prefer Standard Session
Rate Equations for the Study of Intervalle Scattering in Compound Semiconductors C. J. Stanton, University of Florida;* and D. W. Bailey, University of South Carolina - We present solutions to a set of rate equations for the electron dynamics after photoexcitation by a 2.0 eV laser in GaAs and InP. Results obtained, although simpler than full Monte Carlo solutions, closely follow the experimental data and provide insight into intervalley scattering. Calculations shows that the net return time of electrons from the satellite L valleys into the $\Gamma$ valley is not limited by the intervalley scattering rate, but is instead limited by polar optic phonon scattering rate within the $\Gamma$ valley. This shows that the time dependent mobility and luminescence experiments depend on the $L$ valley depopulation rate, which differs from the $L \rightarrow \Gamma$ intervalley scattering rate. Results further suggest that the $\Gamma \rightarrow L$ scattering rate is faster than the polar optic phonon scattering rate.

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Prefer Standard Session
Tunable Pump-Probe Nonlinear Absorption Spectroscopy in AlGaAs D. W. Bailey, University of South Carolina; C. J. Stanton, University of Florida; M. Ulman, L. H. Acioli, J. G. Fujimoto, MIT; * - We report the results of experiments and calculations of the nonlinear transient absorption in AlGaAs systems. In the experiments, the wavelength of the pump and probe pulses can be simultaneously varied from 500 nm to 800 nm. This, for the first time, allows one to selectively study intervalley scattering, since one can tune the region of photoexcited electrons above and below the threshold for scattering into the satellite L valleys. Theoretical calculations based on ensemble Monte Carlo simulations, matrix method techniques and rate equations allow one to decipher the effects of 1) intervalley scattering; 2) density dependence and carrier-carrier scattering; 3) polar optic phonon scattering. Results show these tunable pump-probe experiments are more sensitive and better suited to determining intervalley scattering rates than previous time-dependent mobility and band edge luminescence experiments.

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