

Ultrafast relaxation dynamics of photoexcited carriers in GaAs and related compounds

A. J. Taylor,* D. J. Erskine,† and C. L. Tang

Materials Science Center, Cornell University, Ithaca, New York 14853

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The femtosecond intraband relaxation of hot carriers in GaAs, $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$, and the multiple-quantum-well structure is studied using the equal-pulse optical-correlation technique. An overview of the experimental application of this technique to semiconductors is presented. A detailed theoretical analysis of the coherent-artifact contribution to the transmission-correlation peak in the geometry of parallel copropagating beams and a calculation of the saturable-absorption symmetry coefficients for GaAs are given. The relaxation time of carriers from their initially excited states was measured to be in the range 50–100 fsec for the materials studied. The interpretation of the measured relaxation time in terms of electron and hole response functions is discussed. The relevant scattering processes and rates and the corresponding relaxation times calculated from these rates are given.

1. INTRODUCTION

The study of the ultrafast dynamics of hot carriers in semiconductors is important for the understanding of the basic physics of scattering processes in semiconductors and has motivated many recent experimental optical studies.^{1–5} Moreover, it is crucial to the design of high-speed electronic devices in which the spatial extent approaches the mean free path of the carriers so that the carrier distribution is no longer thermal. We present here a review of our studies^{6–8} of the femtosecond intraband relaxation dynamics of hot carriers in GaAs and related materials using optical techniques. We have measured the initial relaxation time T_r , characterizing the isotropic depopulation of carriers from their initially photoexcited levels as a function of photoexcited carrier density in GaAs, $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$, and a GaAs/AlGaAs multiple quantum-well (MQW) structure at room temperature.

Although our previous papers have discussed mainly the experimental technique and results, we emphasize here the theoretical foundation of the technique and the interpretation of our measurements. We present in Section 2 a brief overview of the application of the equal-pulse technique⁹ to the study of semiconductors. In Section 3, a theoretical background for the experiment is provided: The band structure of GaAs and related materials is described and a detailed analysis of the transmission-correlation peak including the contribution of a coherent artifact is presented. This is the first reported detailed analysis of the coherent artifact contribution in the geometry of copropagating beams and calculation of the saturable-absorption symmetry coefficients for $\text{Al}_{1-x}\text{Ga}_x\text{As}$. In Section 4, we describe the interpretation of the data, including the determinations of T_r and its relation to the behavior of the conduction and valence carrier distributions. In Section 5, we address the issue of attributing T_r to a specific scattering process by itemizing the relevant scattering mechanisms and then estimating their rates.

2. EXPERIMENT

The equal-pulse correlation technique is based on the saturation effect in the transmission of two laser pulses through

a thin sample. The two pulses, which are orthogonally polarized, collinearly propagating, and equal in energy, are focused on the sample with an intensity such that saturable absorption occurs. Their combined, time-averaged, transmitted flux is measured as a function of temporal delay τ . This transmitted flux reaches a peak at $\tau = 0$ and reduces to a background value when τ becomes much greater than the relaxation time of the absorption process and the laser pulse width. We call this peak a transmission correlation peak (TCP). In general, it consists of an incoherent portion (TCP_i), whose shape yields information on the relaxation processes that depopulate the photoexcited state, and a coherent artifact (CA), whose shape contains no such information. Figure 1 shows a typical experimental TCP scan for an AlGaAs sample where the time-averaged transmitted flux of both pulses versus τ is displayed. An autocorrelation (AC) of the laser pulse is also shown.

The equal-pulse correlation technique is particularly useful when studying processes that occur on time scales comparable with or faster than the pulse width (i.e., intraband relaxation) in the presence of much slower processes (such as interband relaxation), since the TCP corresponding to the fast decay process is then clearly revealed as a symmetrical, narrow peak on the flat background, which corresponding to the slow decay. After subtraction of the CA from the TCP, the remaining narrow peak is then proportional to a convolution of the AC and a double-sided exponential with decay time T_r corresponding to the fast process. In contrast, when the conventional pump-probe technique is used,¹⁰ fast processes are obscured by the step in transmission at $\tau = 0$ and by the long decay. A fast decay is revealed mainly through a modification of the rise of the initial step, making it difficult to determine T_r for the fast process.

The experimental arrangement for measuring the TCP is shown in Fig. 2. A passively mode-locked ring laser operating in the colliding-pulse mode produces a train of 90-fsec pulses at 612 nm and at a repetition rate of 10^8 Hz. The pulse train is divided into two arms of orthogonal polarization and equal power by the half-wave plate and the polarizing beam splitter. After retroreflection from the corner-cube prisms, the two arms are recombined collinearly. The time delay between the

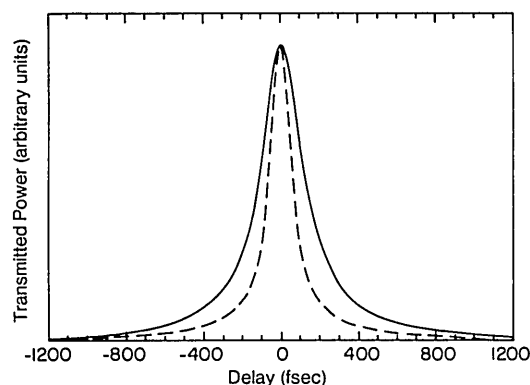


Fig. 1. The solid curve is a TCP for a sample of AlGaAs, and the dashed curve is an AC of the laser pulse.

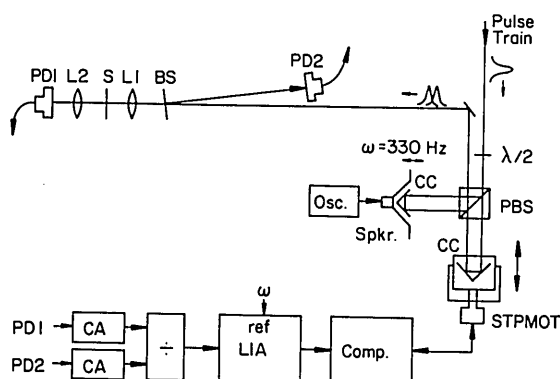


Fig. 2. Optical setup for the equal-pulse correlation technique. PBS, BS, 50% polarizing and 4% ordinary beam splitters, respectively; $\lambda/2$, half-wave plate; CC, retroreflecting corner cubes; SPKR, speaker; STPMOT, stepping-motor-driven linear actuator; PD1, PD2, P-I-N photodiodes. L1, L2, 40X and 20X microscope objectives, respectively; S, sample; CA, current amplifier; \div , dividing circuit; LIA, lock-in amplifier; Comp., computer.

two arms, τ , is determined by the instantaneous positions of the speaker and the linear translator on which the corner cubes are mounted. The recombined pulse train is focused onto the sample by lens L1. The transmitted light is collected by lens L2, and the time-averaged transmitted flux is detected by photodiode PD1. To correct for fluctuations in the laser power during a scan, a portion of the pulse train is reflected by a beam splitter onto PD2. The two photodiodes are connected to current amplifiers, the outputs of which are divided to yield a normalized signal. The resulting signal is digitized and stored by computer for subsequent numerical analysis.

We use a derivative dither technique to improve the signal-to-noise ratio. The speaker corner cube is dithered at a frequency $\omega = 330$ Hz with an excursion of 15 fsec (about 10% of the TCP width). For each value of the average time delay set by the linear translator, the lock-in amplifier detects the component of the signal at ω . A scan is made by recording the amplitude of this component as a function of average delay. Since the dither amplitude is small compared with the width of the TCP, the amplitude of the ω component is proportional to the derivative of the TCP with respect to τ . After the scan is complete, it is integrated numerically to yield the TCP versus τ , as is shown in Fig. 1. The AC is taken using the same experimental setup and method, with an ordinary beam splitter replacing the polarizing one, a 75- μ m-thick ammonium dihydrogen phosphate (ADP) crystal in place of the

sample, and a photomultiplier tube detecting the generated second harmonic light.

Results on three semiconductor samples are reported in this paper: (1) a layer of undoped GaAs 0.3 μ m thick cladded by 0.15- μ m layers of transparent (to 612-nm light) $\text{Al}_{0.6}\text{Ga}_{0.4}\text{As}$, (2) a 0.25- μ m-thick uncladded layer of $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$, and (3) a MQW structure consisting of five 15-nm-thick undoped GaAs wells between 70-nm $\text{Al}_{0.7}\text{Ga}_{0.3}\text{As}$ barriers. All the samples were grown by the conventional low-pressure modified chemical-vapor deposition process, and the substrates were removed in a 375- μ m circle by chemical etching.

3. THEORY

A. Band Structure of GaAs and Related Materials

Figure 3 displays a schematic of the band structures of GaAs, revealing the heavy-hole, light-hole, and split-off valence bands as well as portions of the conduction band along a (111) direction in k space. The conduction band has three relevant minima: the central valley or Γ valley at $k = 0$, the L valley at $k \sim 10^8 \text{ cm}^{-1}$ in the (111) direction, and the X valley (not shown) at $k \sim 10^8 \text{ cm}^{-1}$ in the (100) direction. Although there are no direct 2.02-eV transitions to the L or X valley, these outer valleys participate in important scattering processes. The three direct transitions for 2.02-eV photons in GaAs are indicated in Fig. 3 by the vertical lines. These transitions originate from the heavy-hole (h), light-hole (l), and split-off (s) valence bands. The density of states in the optically coupled regions (OCR's) for the h and l transitions are nearly equal, whereas the number of levels in the OCR of the split-off band is estimated to be 15% of the total number of levels in all three OCR's.

The band structure of $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$ is also displayed in Fig. 3. In AlGaAs the heavy- and light-hole conduction-band OCR's are much closer to the bottom of the central valley than in GaAs (0.17 and 0.10 eV versus 0.51 and 0.45 eV, respectively), which affects the magnitude of some scattering rates. As in GaAs, the densities of states in the OCR for the heavy- and light-hole transitions are roughly equal; however, in AlGaAs there is no 2.02-eV transition from the split-off band owing to the large band gap. Table 1 lists the values of various parameters¹¹ for the band structures of GaAs and AlGaAs.

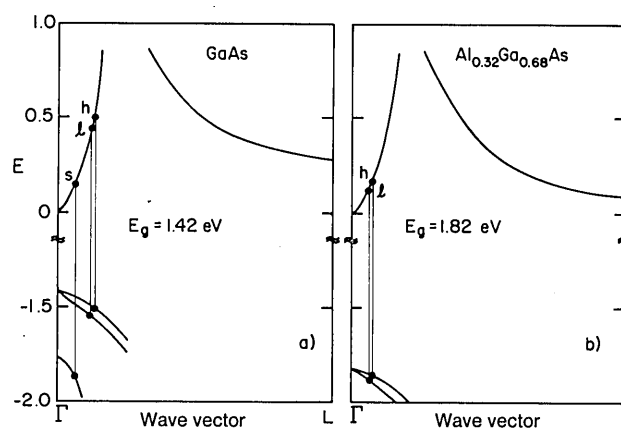


Fig. 3. Band structures of a) GaAs and b) $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$, showing the three valence bands and the central and L valleys of the conduction band. h , l , and s mark the levels optically coupled by the allowed transitions for a 2.02-eV photon from the heavy-hole, light-hole, and split-off valence bands, respectively, to the conduction band.

Table 1. Band-Structure Parameters^a

Parameters	GaAs	AlGaAs
Mass ^b		
$m_{\Gamma h}$	0.5	0.6
$m_{\Gamma s}$	0.145	same ^e
$m_{\Gamma c}$	0.063	0.09
m_L	0.222	same
m_X	0.58	same
Energy (eV) ^c		
E_g	1.42	1.82
$\Delta_{\Gamma L}$	0.29	0.09
$\Delta_{\Gamma X}$	0.48	0.14
Nonparabolicities ^d		
α_{Γ}	0.62	same
α_L	0.46	same
α_X	0.20	same

^a See Ref. 11.^b $m_{\Gamma h, \Gamma s, \Gamma c, L, X}$ are the masses in units of the free-electron mass of the carriers in the heavy and split-off valence bands, in the central-valley conduction band, and in the L and X valleys of the conduction band.^c E_g is the band gap. Δ_{ij} are the energy differences between the minima of the various valleys.^d α_i is the nonparabolicity of the various valleys defined by Eq. (28).^e "Same" means that a value appropriate to $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$ could not be found and that the GaAs value was assumed.

B. Transmission Correlation Peak

To describe the TCP, we use the formalism of Ref. 12, in which the density-matrix equations are iteratively solved for the three-level system consisting of two levels and a reservoir that describes a saturable absorber interacting with an electromagnetic field to calculate the resultant third-order nonlinear polarization. For propagation in the y direction the total incident electric-field envelope has the form

$$\mathbf{E}(t, r) = \exp(iky) \sum_j E_j(t) \hat{e}_j \quad (1)$$

and results in a third-order nonlinear polarization whose envelope is given by

$$\mathbf{P}^{(3)}(t, r) \propto \exp(iky) \sum_{ijkl} \hat{e}_i E_j(t) \times \int_0^\infty dw E_k^*(t-w) E_l(t-w) A_{ijkl}(t-w). \quad (2)$$

A_{ijkl} is the impulse response of the third-order susceptibility for the combination of electric fields $E_i^* E_j E_k^* E_l$:

$$A_{ijkl}(t) = \{Y_{ij} Y_{kl} [1 - \exp(-t/T_0)] + Y_{ijkl} \exp(-t/T_0)\} G(t). \quad (3)$$

$G(t)$ is the impulse response of the population of the photoexcited state. [Note that T_r is the relaxation time describing the initial decay of $G(t)$]. T_0 is the orientational diffusion time that characterizes the decay of the photoexcited anisotropy of the material. Y_{ij} and Y_{ijkl} are coefficients, derived from projection integrals of the transition-matrix elements, which describe the symmetry of the saturable-absorption process.

If the excited-state wave functions ϕ_c^γ are N_2 -fold degenerate wave functions, with γ labeling the degeneracy parameter, and the ground-state wave functions ϕ_v^δ are N_1 -fold degenerate, with δ labeling the degeneracy, then the transition-matrix element $\mathbf{r}_{cv}^{\gamma\delta}$ is defined as

$$\mathbf{r}_{cv}^{\gamma\delta} = \langle c\gamma | \mathbf{r} | v\delta \rangle. \quad (4)$$

The Y parameters can then be written in terms of $\mathbf{r}_{cv}^{\gamma\delta}$ in the following manner:

$$Y_{ij} Y_{kl} = 9/(32\pi^2 N_1) \sum_{\gamma=1}^{N_2} \sum_{\beta=1}^{N_2} \sum_{\delta=1}^{N_1} \sum_{\alpha=1}^{N_1} \times (\int d\Omega \mathbf{r}_{cv}^{\gamma\delta*} \cdot \hat{e}_i^* \mathbf{r}_{cv}^{\gamma\delta} \cdot \hat{e}_j) \times (\int d\Omega [\mathbf{r}_{cv}^{\gamma\alpha*} \cdot \hat{e}_k^* \mathbf{r}_{cv}^{\gamma\alpha} \cdot \hat{e}_l + \mathbf{r}_{cv}^{\beta\delta*} \cdot \hat{e}_k^* \mathbf{r}_{cv}^{\beta\delta} \cdot \hat{e}_l]), \quad (5a)$$

$$Y_{ijkl} = 9/(8\pi^2 N_1) \sum_{\gamma=1}^{N_2} \sum_{\beta=1}^{N_2} \sum_{\delta=1}^{N_1} \sum_{\alpha=1}^{N_1} \times \int d\Omega [(\mathbf{r}_{cv}^{\gamma\delta*} \cdot \hat{e}_i^* \mathbf{r}_{cv}^{\gamma\delta} \cdot \hat{e}_j) \times (\mathbf{r}_{cv}^{\gamma\alpha*} \cdot \hat{e}_k^* \mathbf{r}_{cv}^{\gamma\alpha} \cdot \hat{e}_l + \mathbf{r}_{cv}^{\beta\delta*} \cdot \hat{e}_k^* \mathbf{r}_{cv}^{\beta\delta} \cdot \hat{e}_l)]. \quad (5b)$$

Our definition of the Y parameters differs from that of Ref. 12 since we need to consider the degeneracies that occur in the conduction and valence bands in GaAs. This extension to the degenerate case follows simply from the density-matrix formalism. Note also that we use the matrix elements of \mathbf{r} and not those of its unit vector in the above definitions, since the extension to the degenerate case is then more straightforward; however, the Y parameters are no longer dimensionless.

In the small-signal regime, the induced change in absorption,¹³ and hence the induced change in transmitted intensity, ΔI , is given by

$$\Delta I = -\text{Im}[\mathbf{E}^*(t) \cdot \mathbf{P}^{(3)}(t)]. \quad (6)$$

Since the TCP is the time-averaged change in the transmitted light as a function of delay τ between two input pulses, it is found by combining expressions (2) and (6) that

$$\text{TCP}(\tau) \propto \sum_{ijkl} \int_0^\infty dw A_{ijkl}(w) \times \int_{-\infty}^\infty E_i^*(t) E_j(t) E_k^*(t-w) E_l(t-w) dt. \quad (7)$$

Experimentally, the two input pulses are equal in intensity, collinearly propagating, and orthogonally polarized (o). In this case only six terms are nonzero in expression (7):

$$\begin{aligned} \text{TCP}_o(\tau) \propto & \int_0^\infty dw \left\{ A_{xxzz}(w) \right. \\ & \times \int_{-\infty}^\infty dt [E^*(t) E(t) E^*(t-w+\tau) E(t-w+\tau)] \\ & + A_{zzxx}(w) \int_{-\infty}^\infty dt [E^*(t+\tau) E(t+\tau) \\ & \times E^*(t-w) E(t-w)] + A_{zzzz}(w) \\ & \times \int_{-\infty}^\infty dt [E^*(t) E(t+\tau) E^*(t-w+\tau) E(t-w)] \\ & + A_{zzxz}(w) \int_{-\infty}^\infty dt [E^*(t+\tau) E(t) E^*(t-w) \\ & \times E(t-w+\tau)] + \exp(-2i\omega\tau) A_{zzxz}(w) \\ & \times \int_{-\infty}^\infty dt [E^*(t) E(t+\tau) E^*(t-w) E(t-w+\tau)] \\ & \left. + \exp(2i\omega\tau) A_{zzxz}(w) \int_{-\infty}^\infty dt [E^*(t+\tau) E(t) \right. \\ & \left. \times E^*(t-w+\tau) E(t-w)] \right\}. \quad (8) \end{aligned}$$

The last two terms in expression (8) oscillate with τ as $\exp(2i\omega\tau)$ and therefore are seen only if the experiment resolves fringes separated by 1 fsec (or $0.15\text{-}\mu\text{m}$ steps in a delay line). Although the existence of these fringes has been identified experimentally,¹⁴ our setup is not sensitive to them, since we dither over a $\pm 15\text{-fsec}$ interval at each position of the delay line. If these fringing terms are averaged out to zero by experimental conditions, then expression (8) becomes

$$\text{TCP}_o(\tau) \propto \int_0^\infty d\omega A_{xxzz}(\omega)[\text{AC}(\tau - \omega) + \text{AC}(\tau + \omega)] \\ + \int_0^\infty d\omega A_{zzzz}(\omega)[\zeta(\omega, \tau) + \zeta(\omega, -\tau)], \quad (9)$$

where the symmetry properties of the Y parameters¹² have been used. $\text{AC}(\tau)$ is the AC of the pulse intensity envelope, and $\zeta(\omega, \tau)$ is given by

$$\zeta(\omega, \tau) = \int_{-\infty}^\infty dt E^*(t)E(t + \tau)E^*(t + \tau - \omega)E(t - \omega). \quad (10)$$

An expression for the TCP when the input pulses have parallel polarizations (p) is

$$\text{TCP}_p(\tau) \propto \int_0^\infty d\omega A_{zzzz}(\omega)[\text{AC}(\tau - \omega) + \text{AC}(\tau + \omega)] \\ + \int_0^\infty d\omega A_{zzzz}(\omega)[\zeta(\omega, \tau) + \zeta(\omega, -\tau)]. \quad (11)$$

[Again fringing terms proportional to $\exp(2i\omega\tau)$ are assumed to have been averaged to zero by the experimental conditions.]

C. Coherent and Incoherent Contributions to the Transmission-Correlation Peak

The first term in expressions (9) and (11) represents the incoherent contribution [$\text{TCP}_i(\tau)$] to the saturation process. This is the term that provides information on the relaxation of the populations of the photoexcited state. It is proportional to the convolution of the AC and the response function of the photoexcited state. The second term in expressions (9) and (11) is the CA contribution to the TCP. $\text{CA}(\tau)$ provides no information on the relaxation of the photoexcited state, as it is nonzero only when τ is less than the coherence time of the pulse. If the relevant relaxation process of the material has a decay constant much longer than the laser pulse width, then the CA will not present a problem, since it will simply appear as a spike at $\tau = 0$. However, for processes that are faster than the pulse width, such as intraband relaxation, the amplitude of the CA at $\tau = 0$ as well as its shape must be determined. The CA can then be subtracted from the total TCP, yielding TCP_i , from which a relaxation time may be extracted.

If the laser pulses are bandwidth limited, then the shape of the CA will be approximately the same as the AC. However, the shape of the CA can be more accurately derived from a measurement of the TCP, in which the sample is a thin jet of an organic dye in solution. The central narrow peak in such dyes as Nile blue in ethylene glycol has been determined to be almost completely ($>90\%$) due to the CA.^{8,14} Since the shape of the CA depends mainly on the coherence properties of the pulse and only weakly on material parameters, we can

use this CA from organic-dye molecules to approximate the CA for other materials such as semiconductors.

We define the fractional amount of CA (FCA) at $\tau = 0$ for either parallel (p) or orthogonally (o) polarized input beams as

$$\text{FCA}_{p,o} = \text{CA}(0)_{p,o}/\text{TCP}(0)_{p,o}. \quad (12)$$

An examination of expressions (3), (11), and (12) reveals that $\text{FCA}_p = 1/2$. FCA_o depends on the Y parameters and on T_o . Since $Y_{ij} = 0$ if $i \neq j$, expressions (3), (9), and (10) reveal that, in the orthogonal configuration, the CA is negligible when the absorption properties of the material are polarization independent ($Y_{zzzz} = 0$) or when T_o is much shorter than both the pulse width and the faster energy relaxation process. A simple expression for FCA_o can be derived when the following assumptions are made: (1) The response function is of the form $G(t) = \exp(-t/T_r)$, (2) T_o and T_r are less than the pulse width, and (3) the electric-field envelope is real and temporally Gaussian:

$$\text{FCA}_o = (T_o/T_r) Y_{zzzz}/[(T_o/T_r)(Y_{zzzz} + Y_{xxzz}) + Y_{zz}^2]. \quad (13)$$

Therefore to estimate FCA_o one needs the Y parameters that can be calculated theoretically and the ratio T_o/T_r that can be found from a measurement of the ratio R defined as

$$R = \text{TCP}(0)_p/\text{TCP}(0)_o. \quad (14)$$

Using the approximations listed above, R becomes

$$R = 2[Y_{zz}^2 + Y_{zzzz}(T_o/T_r)] / \\ \times [Y_{zz}^2 + (Y_{zzzz} + Y_{xxzz})(T_o/T_r)], \quad (15)$$

which we can solve for T_o/T_r in terms of R as

$$T_o/T_r = Y_{zz}^2(2 - R)/[R(Y_{zzzz} + Y_{xxzz}) - 2Y_{zzzz}]. \quad (16)$$

Using Eqs. (13) and (16), the calculated values of the Y parameters, and the experimentally determined value of R , the amplitude of the CA contribution to the TCP can be determined. Assuming that the shape of the CA can be approximated by the dye TCP, the CA can then be subtracted from the total TCP. The remaining TCP_i can then be analyzed to determine the relaxation times of the system. Independent of the above assumptions, we note that when $T_o = 0$, $R = 2$ and no CA is present. Conversely, when $T_o = \infty$, $R = 2Y_{zzzz}/(Y_{zzzz} + Y_{xxzz})$ and there is a maximum FCA_o of $Y_{zzzz}/(Y_{zzzz} + Y_{xxzz})$.

D. Y Parameters

To calculate the Y parameters using Eqs. (5), we need the transition-matrix elements and hence the wave functions. Kane¹⁵ derives the wave functions for direct-gap semiconductors with a zinc-blende structure by using a $\mathbf{k} \cdot \mathbf{p}$ perturbation approach and taking into account the spin-orbit interaction. There are six doubly degenerate valence-band states, corresponding to the heavy-hole (h), light hole (l), and split-off (s) valence bands, and two degenerate conduction (c) band states. In the body-fixed coordinate system, where \mathbf{k} is in the z direction, these wave functions have the following form:

Table 2. Relevant Parameters for the 2.02-eV Transitions in GaAs and Al_{0.32}Ga_{0.68}As

Transitions	Parameters					
	k (cm ⁻¹)	E_c (eV)	E_v (eV)	Δ (eV)	E_g (eV)	P (eV/cm)
GaAs				0.34	1.42	9.58×10^{-8}
Heavy hole	1.06×10^7	1.93	-0.086			
Light hole	0.97×10^7	1.87	-0.152			
Split off	0.52×10^7	1.57	-0.45			
AlGaAs				0.31	1.82	9.00×10^{-8}
Heavy hole	0.66×10^7	1.99	-0.028			
Light hole	0.51×10^7	1.92	-0.080			

Table 3. Wave-Function Coefficients for the Possible 2.02-eV Transitions in GaAs and Al_{0.32}Ga_{0.68}As

Transitions	Bands	Coefficients		
		a_i	b_i	c_i
GaAs				
Heavy hole	Conduction	0.91	0.032	0.42
Light hole	Valence	0.13	-0.96	-0.23
Light hole	Conduction	0.91	0.032	0.40
Split off	Valence	-0.22	-0.55	0.80
Split off	Conduction	0.97	0.024	0.27
AlGaAs				
Heavy hole	Conduction	0.97	0.017	0.25
Light hole	Valence	0.15	-0.78	-0.61
Light hole	Conduction	0.97	0.016	0.24

Table 4. Transition-Matrix Element $r_{cv}^{\gamma\delta}$ for the Various Valence- to Conduction-Band Transitions for the Direct-Gap Semiconductors Described by the Wave Functions of Eqs. (17) and (18)^a

Matrix Elements	Transitions	
	$\gamma = 1/2$	$\gamma = -1/2$
$r_{ch}^{\gamma\delta'}$	$\delta = 3/2$	$ia_c r(\hat{x} + i\hat{y})/6^{1/2}$
	$\delta = -3/2$	0
$r_{cl}^{\gamma\delta'}$	$\delta = 1/2$	$-i\rho r\hat{z}/3^{1/2}$
	$\delta = -1/2$	$i\eta r(\hat{x} - i\hat{y})/6^{1/2}$
$r_{cs}^{\gamma\delta'} = r_{cl}^{\gamma\delta'}$		$-i\rho r\hat{z}/3^{1/2}$

^a r is the reduced matrix element for the transitions. ρ and η are derived from the coefficients a_i , b_i , and c_i by $\rho = a_c c_v - a_v c_c$ and $\eta = a_c b_v - a_v b_c$. l refers to the light-hole transition, s to the split-off transition, and h to the heavy-hole transition. \mathbf{k} is assumed to be in the z direction.

$$\phi_h^{3/2} = [(X + iY)\uparrow]/2^{1/2}, \quad (17a)$$

$$\phi_h^{-3/2} = [(X - iY)\downarrow]/2^{1/2}, \quad (17b)$$

$$\phi_i^{1/2} = a_i[iS\uparrow] + b_i[-(X + iY)\downarrow]/2^{1/2} + c_i[Z\uparrow], \quad (17b')$$

$$\phi_i^{-1/2} = a_i[iS\downarrow] + b_i[(X - iY)\uparrow]/2^{1/2} + c_i[Z\downarrow]. \quad (17b'')$$

The indices $\pm 3/2$ and $\pm 1/2$ refer to M_J at $k = 0$, and the index i refers to the l , s , or c band. a_i , b_i , and c_i are real coefficients, given by

$$a_i = kP(E'_i + 2/3\Delta)/N, \quad (18a)$$

$$b_i = 2^{1/2}\Delta(E'_i - E_G)/3N, \quad (18b)$$

$$c_i = (E'_i - E_G)(E'_i - 2\Delta/3)/N, \quad (18c)$$

where N is the normalization factor, E_G is the gap energy, Δ

Table 5. Y Parameters for Direct Band-Gap Semiconductors Described by the Wave Functions of Eqs. (17) and (18)^a

Y Parameter	Transitions	
	Heavy Hole	Light Hole or Split Off
Y_{zz}	$a_c^2 r ^2/3$	$(\rho^2 + \eta^2) r ^2/3$
Y_{zzzz}	$2a_c^4 r ^4/15$	$(3\rho^4 + 2\eta^4 + 2\rho^2\eta^2) r ^4/15$
Y_{xxxx}	$a_c^4 r ^4/10$	$(2\rho^4 + 3\eta^4 + 8\rho^2\eta^2) r ^4/30$
Y_{zzxz}	$a_c^4 r ^4/10$	$(2\rho^4 + 3\eta^4 - 2\rho^2\eta^2) r ^4/30$

^a r is the reduced matrix element for the transitions. ρ and η are derived from the coefficients a_i , b_i , and c_i by $\rho = a_c c_v - a_v c_c$ and $\eta = a_c b_v - a_v b_c$, where $v = 1$ for the light-hole transition and $v = s$ for the split-off transition.

Table 6. Y Parameters^a for the Possible 2.02-eV Transitions in GaAs and Al_{0.32}Ga_{0.68}As

Transitions	Y Parameters			
	Y_{zz}	Y_{zzzz}	Y_{xxxx}	Y_{zzxz}
GaAs				
Heavy hole	0.28	0.091	0.069	0.069
Light hole	0.28	0.087	0.074	0.056
Split off	0.33	0.135	0.093	0.027
Total	0.29	0.092	0.073	0.060
AlGaAs				
Heavy hole	0.31	0.118	0.089	0.089
Light hole	0.32	0.105	0.104	0.029
Total	0.31	0.112	0.096	0.059

^a The total Y parameter for the 2.02-eV transition can be found by averaging each Y parameter over the possible transitions, weighting each one by the density of states squared. Y_{zz} is given in units of $|r|^2$, and Y_{zzzz} , Y_{xxxx} , and Y_{zzxz} are given in units of $|r|^4$, where r is the reduced matrix element for the transitions.

is the spin-orbit splitting, and E'_i is the energy of the state, given by $E_i - \hbar^2 k^2/2m$. The symbols \uparrow and \downarrow refer to spin quantum numbers of $\pm 1/2$. The function S has the symmetry properties of the atomic s functions under the operations of the tetrahedral group, while X , Y , and Z have the symmetry properties of the atomic p functions x , y , and z under that group. In Table 2 we present the relevant parameters for the 2.02-eV transitions in GaAs and AlGaAs, and in Table 3 we present the wave-function coefficients a_i , b_i , and c_i .

The matrix elements $r_{cv}^{\gamma\delta'}$ for all transitions from the valence band to the conduction band in the body-fixed coordinate system, where \mathbf{k} is in the z direction, are tabulated in Table 4. The laboratory-fixed coordinate system (defined by the \mathbf{E} field) is specified with respect to the body-fixed frame by the usual Euler angles¹⁶ α and β . (γ can be set equal to zero since in the body-fixed frame only the z direction is relevant.) The new matrix elements in this frame, $r_{cv}^{\gamma\delta}$, can be

found from a rotation:

$$\mathbf{r}_{cv}^{\gamma\delta} = M \mathbf{r}_{cv}^{\gamma\delta'}, \quad (19)$$

where M is a rotation matrix¹⁶ of the form

$$M = \begin{pmatrix} \cos \beta \cos \alpha & -\sin \alpha & \sin \beta \cos \alpha \\ \cos \beta \sin \alpha & \cos \alpha & \sin \beta \sin \alpha \\ -\sin \beta & 0 & \cos \beta \end{pmatrix}. \quad (20)$$

Using Eqs. (5) and (20) and the matrix elements of Table 4, the calculation of the Y parameters is straightforward but tedious. The resulting expressions are tabulated in Table 5 for the three possible transitions.

The calculated values of the Y parameters for the 2.02-eV transition in GaAs and AlGaAs are presented in Table 6. Total Y parameters for the 2.02-eV transition can be found by averaging each Y parameter over the possible transitions and weighting each transition by the density of states squared. These values are also displayed in Table 6.

4. INTERPRETATION OF DATA

A. Determination of Relaxation Times

From the values of the total Y parameters for the 2.02-eV transition, we note that if $T_o = \infty$, then $R = 1.38$ (1.45), resulting in a maximum FCA of 45% (38%) in GaAs (AlGaAs). Experimentally, in both GaAs and AlGaAs we measure $R = 1.75$, which implies [Eqs. (13) and (16)] that $T_o/T_r = 0.43$ in GaAs and $T_o/T_r = 0.51$ in AlGaAs and that FCA = 17% for both materials. When this CA contribution is subtracted from the total TCP, the remaining TCP_i for the fast process is approximately 30% wider than the corresponding total TCP.

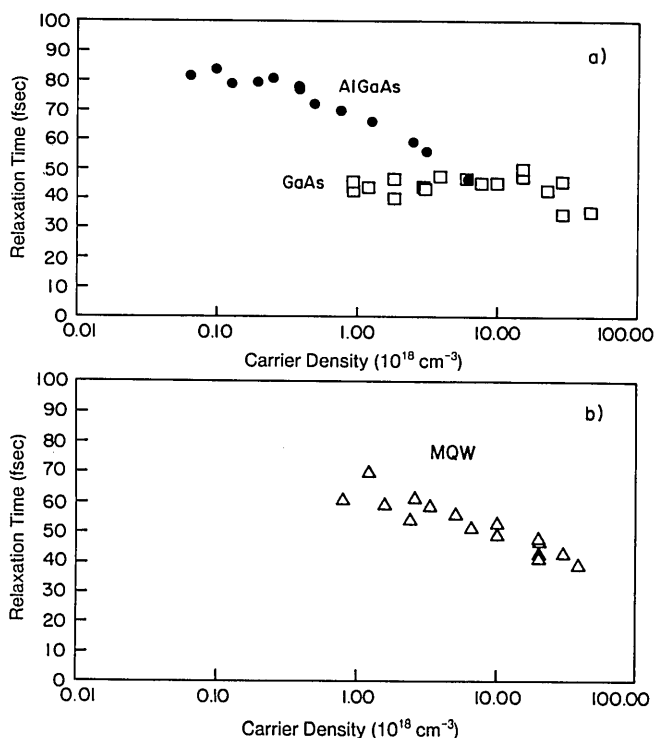


Fig. 4. Relaxation time T_r versus photogenerated carrier density for a) GaAs and AlGaAs and b) MQW.

The values for T_r are determined by deconvolving the measured laser pulse AC from the measured TCP_i. These values for T_r are plotted as a function of photoexcited carrier density n for GaAs, AlGaAs, and the MQW in Fig. 4. At low powers, where carrier-carrier scattering is not expected to be important, T_r is determined by phonon-scattering rates. Thus these data indicate that the phonon rates are greater for GaAs than for AlGaAs. As the input power increases, we expect the carrier-density-dependent scattering rate to grow in importance and compete with the existing photon rates. This is consistent with the downward slope of the curve for AlGaAs. For GaAs, the low-power scattering rate is already large, and the effect of the carrier-density-dependent rate is not seen in the power range studied.

B. Relation of T_r to Carrier Relaxation Times

Since T_r characterizes the initial portion of the decay of the response function, we must first examine its relation to the carrier relaxation times before interpreting the measured values of T_r in terms of the various scattering processes. We need to determine whether T_r characterizes the behavior of the electron or the hole distributions (or a combination of both). In general, the impulse response function $G(t)$ of the saturable-absorption process is due to both electrons occupying the OCR of the conduction band and holes occupying the OCR of the valence band. Thus $G(t)$ is a weighted sum of both the separate response functions for electrons and holes [$G_e(t)$ and $G_h(t)$, respectively]:

$$G(t) = aG_e(t) + (1 - a)G_h(t), \quad 0 \leq a \leq 1. \quad (21)$$

The weighting factor a is linked to the number of levels M_v and M_c in the valence- and conduction-band OCR's, respectively. Let the number of filled and vacant levels in the valence-band OCR be n_v and p_v , such that $n_v + p_v = M_v$, and those for the conduction-band OCR be n_c and p_c , such that $n_c + p_c = M_c$. Then the net change in the number of photons transmitted through a thin layer of material is proportional to $n_v p_c - n_c p_v = M_c M_v - (M_v n_c + M_c p_v)$. Thus if G_e and G_h describe the decay of n_c and p_v , then the response function for the saturable absorption is

$$G(t) \propto M_v G_e(t) + M_c G_h(t), \quad (22)$$

and therefore $a = M_v / (M_c + M_v)$.

M_c and M_v are determined by the widths of the OCR's in the conduction and valence bands, respectively. In the case in which the photon bandwidth is large compared with the material linewidth, conservation of momentum ensures that $M_c = M_v$. In the case in which the material linewidth is comparable with or larger than the photon bandwidth, the widths (in energy) of the OCR's in the conduction and valence bands are comparable. If the valence-band effective mass is much larger than the conduction-band effective mass, then M_v can be larger than M_c . In this case, $1 > a > 1/2$, so that $G(t)$ (and hence T_r) describes the response of the electrons more than the response of the holes.

In general, $G(t)$ may be attributed to either $G_e(t)$ or $G_h(t)$ because of different decay behavior of the two response functions. Given that T_r describes the initial decay of $G(t)$ on a time scale of the order of or faster than the pulse width, then if $G_e(t)$ [$G_h(t)$] decays in a time comparable with the pulse width, the measured TCP will emphasize the convolu-

tion of $G_e(t)$ [$G_h(t)$], if the decay of $G_h(t)$ [$G_e(t)$] is either very much faster or very much slower than $G_e(t)$ [$G_h(t)$].

There are two reasons why the decay behavior of G_e and that of G_h may differ. First, $G_e(t)$ and $G_h(t)$ may decay at different rates because the scattering processes, which in part determine the decay behavior, may be different for the conduction and valence bands. This can be checked by simply evaluating the rates of all the relevant scattering processes. Second, $G_e(t)$ and $G_h(t)$ may be affected to different degrees by a band-filling process. (The partial saturation of the OCR owing to carriers in the OCR after relaxation of the initially photoexcited nonequilibrium distribution is termed band filling.) The effect of band filling is to raise the asymptotic level that $G_h(t)$ and $G_e(t)$ approach, so that the influence of the initial decay, corresponding to T_r , is reduced in G_h or G_e . If the photon bandwidth is much larger than the material bandwidth, then the degree of band filling for the electron and hole distributions will be equal, and this effect will not be relevant. If not, the degree of band filling will be larger in the valence band than in the conduction band, if the effective mass of the holes is much greater than that of the electrons, thus reducing the effective contribution of G_h to the initial decay time T_r .

5. SCATTERING PROCESSES

The scattering processes that are relevant to calculating the time evolution of the carrier distribution on a subpicosecond time scale can be grouped into two classes: processes in which the basic scattering rate is independent of carrier density, such as carrier-phonon scattering, and processes in which the rate is dependent on carrier density, such as carrier-carrier scattering. We shall first discuss the relevant carrier-density-independent processes, which consist of polar optical phonon (POP) scattering, acoustic phonon scattering (ACS), random potential alloy scattering (ALLOY), and intervalley (IV) phonon scattering. The estimated scattering rates of these processes, calculated from the material parameters given in Table 7, are summarized in Table 8 and are used to calculate a value for T_r . This value is compared with the measured value of T_r at low carrier densities, where carrier-density-dependent processes are expected to be less important. Finally, the carrier-density-dependent processes are discussed and their rates are estimated.

Table 7. Parameters^a Used in the Calculation of Scattering Rates

Dielectric Constants	$\epsilon_\infty = 10.88$ $\epsilon_0 = 12.85$
Central-valley and IV phonon energies	$\hbar\omega_{op} = 0.036$ eV $\hbar\omega_{ij} = 0.8\hbar\omega_{op}$ eV
Mass density	$\rho = 5.37$ gm/cm ³
Speed of sound	$v_L = 5.22 \times 10^5$ cm/sec
Deformation potentials	$D_{acs} = 7$ eV $D_{rL} = 1 \times 10^9$ eV/cm $D_{rX} = 1 \times 10^9$ eV/cm
Number of equivalent valleys	$Z_l = 4$ $Z_X = 3$

^a See Refs. 11 and 20.

A. Carrier-Density-Independent Processes

The most important intravalley phonon scattering process is POP scattering. Since the energy of an optical phonon ($\hbar\omega_{op} = 0.036$ eV) is relatively independent of its wave vector, the scattered electron loses or gains energy in units of $\hbar\omega_{op}$. In the POP interaction, the displacement of the lattice by the vibration of the optical phonon can cause an induced polarization. This happens only in polar materials, such as $\text{Al}_x\text{Ga}_{1-x}\text{As}$, in which the optical mode of the phonon directly changes the separation between the gallium and arsenic atoms. Since these atoms do not carry equivalent charges, a polarization is developed, which in turn interacts strongly with the charged carrier.

The expressions for the scattering rates for absorption of a POP, R_{POP}^+ , and for emission of a phonon, R_{POP}^- , which include the effects of nonparabolicity of the conduction band but neglect screening, have been derived by Fawcett *et al.*¹⁷:

$$R_{\text{POP}} = e\zeta(2m\gamma)^{1/2}(1 + 2\alpha E')F_o(E, E') \times \begin{cases} N(\omega_{op}) & \text{absorption} \\ N(\omega_{op}) + 1 & \text{emission} \end{cases}, \quad (23)$$

where

$$F_o(E, E') = (1/C) \left[A \ln \frac{|\gamma^{1/2}(E) + \gamma^{1/2}(E')|}{|\gamma^{1/2}(E) - \gamma^{1/2}(E')|} + B \right], \quad (24)$$

$$A = \{2(1 + \alpha E)(1 + \alpha E') + \alpha[\gamma(E) + \gamma(E')]\}^2, \quad (25)$$

$$B = -2\alpha\gamma^{1/2}(E)\gamma(E')\{4(1 + \alpha E)(1 + \alpha E') + \alpha[\gamma(E) + \gamma(E')]\}, \quad (26)$$

$$C = 4(1 + \alpha E)(1 + \alpha E')(1 + 2\alpha E)(1 + 2\alpha E'), \quad (27)$$

$$\gamma(E) = \hbar^2 k^2 / 2m = E(1 + \alpha E), \quad (28)$$

$$E' = \begin{cases} E + \hbar\omega_{op} & \text{for absorption} \\ E - \hbar\omega_{op} & \text{for emission} \end{cases}, \quad (29)$$

$$e\zeta = me^2\omega_{op}(\epsilon_\infty^{-1} - \epsilon_0^{-1})/\hbar^2, \quad (30)$$

$$N(\omega_{op}) = [\exp(\hbar\omega_{op}/k_B T) - 1]^{-1}. \quad (31)$$

The nonparabolicity coefficient of the valleys (α) is defined by Eq. (28). E is the carrier energy measured from the bottom of the valley, and $e\zeta$ is an effective electric field that determines the carrier coupling to the polar modes. $N(\omega_{op})$ is the occupation probability for a phonon having an energy $\hbar\omega_{op}$ and is found from the Bose-Einstein formula. T is the lattice temperature, which is assumed to be 300 K. (Parameters used for calculating scattering rates can be found in Table 7.)

The expression for $R_{\text{POP}}^{+,-}$ is plotted in Fig. 5 for the parameters of GaAs in the central valley of the conduction band. Note that the rates are independent of carrier energy for energies greater than several optical phonons from the valley floor. The calculated POP scattering rates for the heavy- and light-hole OCR in the conduction and valence bands of GaAs and $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$ at 300 K are summarized in Table 8. Finally, note that the POP rates for holes are larger than those for electrons because the magnitude of the rate goes as $m^{1/2}$.

The other intravalley phonon-scattering process is scattering by acoustic phonons. Unlike the optical phonon, the

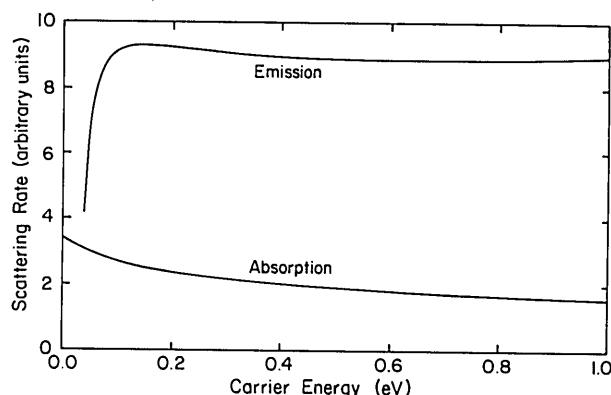


Fig. 5. Scattering rates for emission and absorption of POP's as a function of energy for the conduction band of GaAs. This includes the effects of nonparabolic valleys but neglects screening.

Table 8. Calculated Scattering Rates^a from the Heavy and Light OCR

Type of Scattering	Scattering Rates			
	Electron OCR		Hole OCR	
	Heavy	Light	Heavy	Light
GaAs				
R_{POP}^+	1.7	1.7	5.5	5.5
R_{POP}^-	6.4	6.4	18.5	18.5
R_{ACS}	0.6	0.6	4.7	6.2
R_{ALLOY}	0	0	0	0
R_{IV}	25(12)	19(0)	0	0
AlGaAs				
R_{POP}^+	2.2	2.2	6.8	6.4
R_{POP}^-	7.9	7.9	0	19.8
R_{ACS}	0.5	0.4	3.5	6.3
R_{ALLOY}	1.0	1.0	0.4	0.4
R_{IV}	15(9)	9(0) ^f	0	0

^a Rates are in units of 10^{12} sec^{-1} . $R_{POP}^{+,-}$ is polar optical scattering for absorption (+) and emission (-). R_{ACS} is acoustic and includes emission and absorption of a phonon, R_{ALLOY} is random potential alloy scattering. Note that the latter two rates do not affect the carriers energy but only its momentum; thus they affect only T_0 rather than T_r . R_{IV} is $\Gamma \rightarrow L$ IV deformation-potential scattering by emission and absorption of a phonon using a deformation potential of $1 \times 10^9 \text{ eV/cm}$. The values in parentheses are the $\Gamma \rightarrow X$ rates.

energy of the acoustic phonon is small, making the ACS process approximately elastic in nature. However, the momenta of these phonons can be compared with the momenta of the carriers, so that significant changes in carrier momenta can occur. Fawcett *et al.*¹⁷ give the total scattering rate (absorption and emission) assuming parabolic bands as

$$R_{ACS} = 2^{1/2} D_{ACS}^2 k_B T E^{1/2} m^{3/2} / (\pi \hbar^4 \rho v_L^2), \quad (32)$$

where ρ is the mass density of the material, v_L is the speed of sound, and D_{ACS} is a deformation potential. Generally, the acoustic rate is significantly large only where the effective mass is large, such as for holes. In comparison with the other scattering processes, it is not a significantly large rate for the electron.

Another scattering process, also elastic in nature, is ALLOY. This is only appropriate for the sample of $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$ in which inhomogeneities in its composition occur that are due to random fluctuations of the concentrations of aluminum, gallium, and arsenic atoms. These inhomogeneities can act as scattering centers analogous to impurity scattering centers. Random potential scattering is discussed by Hauser *et al.*¹⁹

and Littlejohn *et al.*,¹⁹ where the scattering rate for a general alloy $A_x B_{1-x} C$ is given as

$$R_{ALLOY} = (3\pi m^{3/2} / 8\hbar^4 2^{1/2}) x(1-x) \times \gamma(E) (1 + 2\alpha E) \Omega |\Delta U|^2 S, \quad (33)$$

where x is the mole fraction of element A, $\Omega = a^3/4$ is the primitive cell volume, and a is the lattice constant. S is a parameter between 0 and 1 that describes the randomness of the alloy. $S = 0$ would describe a perfectly ordered superlattice, and $S = 1$ a completely random alloy. ΔU is the scattering potential. ΔU is taken to be either the difference in energy band gaps between the binary constituents or the difference in electron affinities between the binary constituents. If we take the difference in energy band gaps and attribute 85% to an electron-potential step, as it is usually done in GaAs/AlGaAs heterojunctions, the value for ΔU is 0.37 for electrons and 0.065 eV for holes. The calculated values for R_{ALLOY} , based on these values for ΔU , are given in Table 8.

Both ALLOY and ACS will not significantly affect the value of T_r , since these processes do not change the carrier energy enough to remove it from the OCR. To the extent that the OCR is a spherical shell, scattering processes that change only the carrier's momentum will not remove that carrier from the OCR. (The band structure of GaAs is actually slightly non-spherical, so the OCR is slightly fluted. This is expected to have only a minor effect on the scattering rates.) However, these elastic scattering processes are important in determining the orientational relaxation time T_0 .

In the IV phonon-scattering process an electron scatters from the central valley to one of the outer valleys either by absorption or by emission of a phonon. This process is therefore permitted only when the initial energy of the electron is greater than one optical phonon below the outer-valley minimum. For the L valley in GaAs this energy is greater than 0.3 eV, and in AlGaAs it is greater than 0.1 eV. Because the phonon involved here has a wave vector that is an appreciable fraction of the Brillouin zone, the energy of the optical phonon involved is slightly less than its energy at the zone center ($0.8\hbar\omega_{op}$). This is the most important process in the decay of the electron impulse response. Since there are no outer valleys in the valence band, holes do not participate in IV scattering processes.

Fawcett *et al.*¹⁷ give the IV absorption and emission scattering rates for nonparabolic valleys as

$$R_{IV} = \frac{Z_j m_j^{3/2} D_{ij}}{2^{1/2} \pi \rho \omega_{ij} \hbar^2} E_j^{1/2} G(E_i, E_j) \times \begin{cases} N(\omega_{op}) & \text{absorption} \\ N(\omega_{op}) + 1 & \text{emission} \end{cases}, \quad (34)$$

where E_i is the initial electron energy measured from the initial (i th) valley floor. E_j is the final electron energy measured from the final (j th) valley floor, give or take the IV phonon energy $\hbar\omega_{ij}$:

$$E_j = E_i - \Delta_{ij} \pm \hbar\omega_{ij}, \quad (35)$$

where Δ_{ij} is the energy difference between the floors of the initial and final valleys. Z_j and m_j are the number of equivalent final valleys and the effective mass of the carrier in the final valley. The function $G(E_i, E_j)$ involves the nonparabolicities and is of the order of unity:

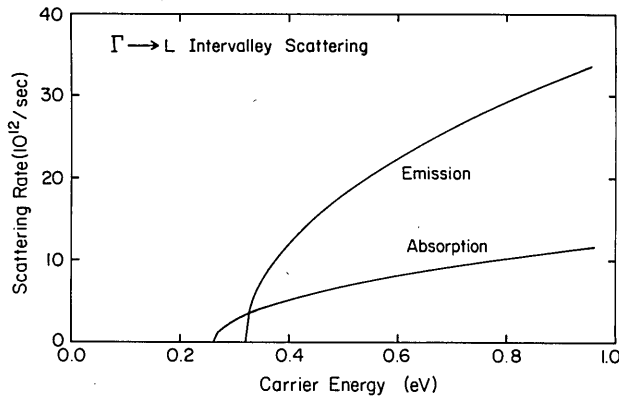


Fig. 6. Scattering rates for emission and absorption of IV phonons, $\Gamma \rightarrow L$, as a function of initial electron energy above the central valley.

$$G(E_i, E_j) = (1 + \alpha_i E_i)(1 + \alpha_i E_i) / [(1 + 2\alpha_i E_i)(1 + 2\alpha_j E_j)]. \quad (36)$$

D_{ij} is the deformation potential specific to the pair of initial and final valleys. We use the value of D_{ij} given by Littlejohn et al.²⁰ Note that this parameter is difficult to determine accurately.

A plot of these rates versus carrier energy for scattering between the central and the L valleys is given in Fig. 6. The energy dependence of the curves is given chiefly by the density of states, which varies as $E^{0.5}$. The values of the $\Gamma \rightarrow L$ and $\Gamma \rightarrow X$ IV rates for the heavy and the light OCR's are tabulated in Table 8 for GaAs and $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$ at 300 K. The $\Gamma \rightarrow X$ rates are enclosed in parentheses. The IV scattering rates from the OCR are larger in GaAs than $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$ because the OCR's are higher above the bottom of the L valley in GaAs. Although both the light and the heavy OCR's are above the L valley, only the heavy OCR is above the X valley, so the light OCR does not participate in $\Gamma \rightarrow X$ scattering. Since the position of the X valley is not known with great accuracy, it is possible that the $\Gamma \rightarrow X$ rate is different from that given here.

Table 8 summarizes the results of all four scattering processes considered for the heavy and the light OCR's. Clearly, the IV process dominates the scattering in the conduction band. In the valence band where there is no IV scattering, POP scattering is the most significant, but it is not so large as the IV rates for the conduction band. Thus the scattering rate for electrons is calculated to be faster than that for holes, owing to the fast IV rates.

Based on the scattering rates given in Table 8 (for IV scattering to the L valley only), the initially excited electron and hole populations should decay with time constants of 30 and 60 fsec, respectively, in GaAs and 40 and 150 fsec, respectively, in AlGaAs. Because of the considerations discussed earlier, we expect T_r to correspond mainly to the decay of the electron population. This yields calculated decay times of 30 fsec in GaAs and 40 fsec in AlGaAs. The use of only the electron response to determine T_r represents the limit $a = 1$ in Eq. (21) that yields a lower bound for T_r . An upper bound for T_r can be found by assuming that the electron and hole populations contribute equally to the response [$a = 0.5$ in Eq. (21)]. In this case, our calculated values for T_r are 40 fsec in GaAs and 80 fsec in AlGaAs. The measured values, at low carrier den-

sity, of 45 and 80 fsec are in good agreement with these calculated estimates.

B. Carrier-Density-Dependent Processes

The values for T_r calculated from carrier-phonon-scattering processes agree well with the measured value of T_r at extreme low carrier densities. However, the variation of the measured data with increasing photoexcited carrier density (Fig. 4) shows that there are carrier-density-dependent processes contributing to T_r that have not yet been taken into account. The relevant carrier-density-dependent processes are the effect of screening on the carrier-phonon scattering rate and carrier-carrier scattering. The latter process manifests different behavior at different carrier densities. At high carrier densities, the collective oscillation of the plasma interactions occur. Free carrier scattering occurs at low carrier densities. Free carrier scattering in turn can be classified as either unlike-particle collisions (electron-hole) or identical-particle collisions (electron-electron, hole-hole).

In general, calculating the effect of these carrier-density-dependent processes on T_r is difficult. Although carrier-density-independent rates permit the determination of the time dependence of the carrier population in the OCR by means of the response or Green's function $G(t)$ (as was assumed to exist in the analyses of the TCP presented in Section 3), for carrier-density-dependent rates, the corresponding rate equations become nonlinear, rendering a response-function approach to the determination of the TCP shape inadequate. As an approximation, a response function is assumed to exist even for carrier-density-dependent processes, decaying with a constant rate that is calculated by using the average carrier density developed under the absorption of the pulse pair. Furthermore, unlike POP or IV scattering, the energy of the scattered carrier is not shifted by a quantum but can fall into a continuum of values, constrained by the conservation of energy and crystal momentum for the scattering particles. Third, theoretical expressions found in the literature concerning these carrier-density-dependent processes almost universally assume a carrier distribution at equilibrium (see, for example, Ref. 21). However, under our experimental conditions, the carrier distribution is highly nonequilibrium- and delta-function-like in character.

Expressions for the free carrier scattering rate as well as for the POP scattering rate involve the screening parameter q_o . Under equilibrium conditions, q_o is given by the well-known formula²¹

$$q_o^2 = 4\pi e^2 n / \epsilon k_B T_c, \quad (37)$$

where T_c is the temperature of the carrier system. This expression for q_o was used in our calculations even though its validity under nonequilibrium conditions has not been established. The problem of screening under nonequilibrium conditions clearly needs further consideration.

An expression for POP scattering that includes screening but ignores the valley nonparabolicity is

$$R_{\text{POP}^{+,-}} = e \zeta(2mE)^{1/2} \left\{ \frac{N(\omega_{\text{op}})}{N(\omega_{\text{op}}) + 1} \times \left[\frac{1}{2} \ln \left(\frac{(q_{\text{max}}^2 + q_o^2)}{(q_{\text{min}}^2 + q_o^2)} \right) + \frac{1}{2} q_o^2 [(q_{\text{max}}^2 + q_o^2)^{-1} + (q_{\text{min}}^2 + q_o^2)^{-1}] \right] \right\}, \quad (38)$$

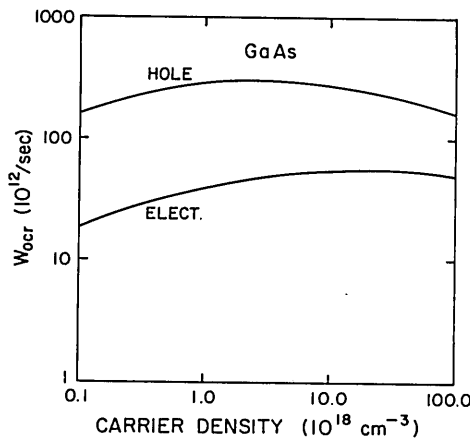


Fig. 7. Calculated free-carrier-carrier rate out of the OCR versus carrier density for GaAs.

where $q_{\max, \min}$ are the limits on the phonon wave vector. For absorption,

$$q_{\max, \min} = k[(1 + \hbar\omega_{\text{op}}/E)^{1/2} \pm 1], \quad (39)$$

with k being the initial carrier wave vector. For emission,

$$q_{\max, \min} = k[1 \pm (1 - \hbar\omega_{\text{op}}/E)^{1/2}]. \quad (40)$$

At a carrier density of 10^{19} cm^{-3} , the calculated effect of screening on POP scattering is to reduce this rate by a factor of about 2 from its unscreened value.

Screening also affects the character of carrier-carrier scattering. The differential cross section for a collision between two free identical particles given by Ridley²² is

$$\begin{aligned} \sigma(\alpha, \mathbf{k}_{cm}) = & U \{ [\mathbf{k}_{cm}^2(1 + \cos \alpha) + q_o^2/4]^{-2} \\ & + [\mathbf{k}_{cm}^2(1 - \cos \alpha) + q_o^2/4]^{-2} \\ & + [\mathbf{k}_{cm}^2(1 + \cos \alpha) + q_o^2/4]^{-1} \\ & \times [\mathbf{k}_{cm}^2(1 - \cos \alpha) + q_o^2/4]^{-1} \}, \quad (41) \end{aligned}$$

where $U = e^4 \mu^2 / \epsilon^2 \hbar^4$, \mathbf{k}_{cm} is the wave vector of the particle in the center-of-mass frame, α is the scattering angle, and μ is the reduced mass. The expression for unlike-particle collision is just the first term of Eq. (41). The effect of screening on carrier-carrier collisions is to increase the predominance of forward angle scattering for small values of q_o/\mathbf{k}_{cm} . As q_o grows, the characteristic scattering angle increases.

In the limit of large q_o , when the screening length $1/q_o$ is larger than the intercarrier spacing, collective oscillations of the plasma become the dominant mode of excitation, and two-body collisions are no longer relevant. The collective oscillations carry quanta of energy $\hbar\omega_p$, where $\hbar\omega_p$ is the plasma frequency. We are not aware of an expression for carrier-plasmon scattering applicable to a monoenergetic carrier distribution.

The effect of free carrier scattering on T_r can be estimated by considering a density of monoenergetic carriers having an energy that places them in the OCR but with a random distribution of wave-vector directions. We calculate the rate of scattering events between carriers that remove carriers from the OCR, averaged over all possible pairs of carriers. Whether a scattering event removes a carrier from the OCR depends on the scattered wave-vector direction and magnitude and on the size of the OCR.

We have numerically calculated this rate for an assumed

size of the OCR, summing over all possible final states, counting only those outside the OCR, and weighing each by the cross section for that event given by Eq. (41). The rates for electron-hole and electron-electron scattering were summed to give the total rate for electrons, and similarly for holes.

The resulting rate of scattering out of the OCR, denoted W_{OCR} , is given in Fig. 7 for electrons and holes in GaAs. The chief result is that W_{OCR} for holes is an order of magnitude larger than that for electrons. This is because W_{OCR} is proportional to the effective mass. The lack of a strong density dependence in the rates is due to the counteractive effects of increased screening and the increased frequency of collisions as the carrier density increases. Calculated W_{OCR} for AlGaAs is similar in its magnitude and density dependence to the results shown in Fig. 7 for GaAs. Although the experimentally measured values for T_r show a density dependence for the AlGaAs sample, but not for the GaAs sample, this calculation of W_{OCR} provides no theoretical reason for the difference in behavior for the two materials.

In the calculation, the resulting value of W_{OCR} at low carrier density is sensitive to the size of the OCR assumed. The value used here, 40 meV for the uncertainty in the transition energy, was based on a semiconductor state lifetime of ~ 50 fsec, together with the measured bandwidth of the laser light. The calculated result, that W_{OCR} for holes corresponds to a 10-fsec relaxation time, indicates a lack of self-consistency in the calculation of W_{OCR} . For if the semiconductor-state lifetime is indeed 10 fsec, as this calculation suggests, then a much broader OCR would result. This in turn would yield a relaxation time for holes much slower than 10 fsec, since more scattered states will fall inside rather than outside the OCR.

This lack of self-consistency indicates that a more comprehensive treatment of the entire problem of carrier-carrier scattering must be found before detailed quantitative comparison between theory and experiment can be made. However, it is likely that the order-of-magnitude faster scattering rate for holes than for electrons would also result from such a calculation. As discussed in Section 4, this result implies that the experimentally measured value for T_r can be attributed to decay of electron distribution rather than to that of hole distribution.

6. CONCLUSIONS

In conclusion, we have presented our results on the femtosecond relaxation of hot carriers in GaAs, $\text{Al}_{0.32}\text{Ga}_{0.68}\text{As}$, and a MQW structure in which we have measured relaxation times of carriers from their initially excited states to be in the range of 50–100 fsec. We have described the theoretical foundations of our experimental technique, the equal-pulse correlation technique, with a detailed analysis of the CA. This is the first reported analysis of the CA contribution in the geometry of parallel copropagating beams and the first reported calculation of the saturable-absorption symmetry coefficients for GaAs and related materials. We have described the interpretation of the measured relaxation time in terms of the decay of electron and hole response functions and concluded that it corresponds mainly to the decay of electron population. Relevant carrier-density-independent scattering processes have been described and their corresponding rates calculated. The IV phonon-scattering process is found to be the dominant

scattering process. Using these calculated scattering rates and the electron response only to determine the relaxation time yielded relaxation times of 30 fsec in GaAs and 40 fsec in AlGaAs in this low-carrier-density limit, in reasonable agreement with our data. Carrier-density-dependent processes have been discussed, and the corresponding rates are estimated wherever possible. The carrier-density dependence of our data is, however, not yet completely understood.

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* Permanent address, AT&T Bell Laboratories, Crawford Hill Laboratory, Holmdel, New Jersey 07733.

† Permanent address, Department of Physics, University of California, Berkeley, California 94720.

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