

Femtosecond Relaxation of Photoexcited Nonequilibrium Carriers in $\text{Al}_x\text{Ga}_{1-x}\text{As}$

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Optical saturation of highly excited states in the conduction band of a semiconductor and the subsequent extremely fast energy relaxation due to carrier-carrier and optical-phonon scattering at room temperature are observed. The corresponding measured lifetime for states 160 meV above the band edge in $\text{Al}_{0.34}\text{Ga}_{0.66}\text{As}$ is in the range of 80 to 30 fs depending upon the photoexcited carrier density.

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There has recently been a great deal of interest in the dynamics of hot carriers following photoexcitation by picosecond light pulses in semiconductors.¹⁻⁴ It is generally thought that at low temperatures carriers generated in states far above the band edge relax first by carrier-carrier scattering and reach a quasiequilibrium distribution at a carrier temperature far above the lattice temperature in less than a picosecond. This hot carrier distribution subsequently cools down to the lattice temperature in a few to tens of picoseconds, depending on temperature, through the emission of phonons. Previous studies have concentrated mainly on the dynamics of the carriers during this second stage of phonon cooling at low temperatures where the time constants involved are on the order of 0.5 ps or more. There has been no direct observation of the relaxation from the initially photoexcited nonequilibrium carrier distribution at room temperature where the relaxation time is in the femtosecond time domain and both optical-phonon (OP) and carrier-carrier scattering could be important. We report here the observation of the saturation of the conduction-band energy shells in \vec{k} space and the measurement of the relaxation time of the conduction-band electrons out of the initially photoexcited states. This relaxation time is clearly of fundamental importance because it limits the intrinsic lifetime of the corresponding Bloch states in the conduction band. It is also important for the understanding of, for example, high-speed GaAs devices and laser annealing processes. In this Letter, we report the direct measurement of this

relaxation time in $\text{Al}_{0.34}\text{Ga}_{0.66}\text{As}$ at room temperature using a femtosecond laser.⁵

Our measurement is based upon a study of the saturation effect on the transmission characteristics of a thin sample of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ using a new technique for measuring femtosecond excited-state relaxation times called the equal-pulse correlation technique. In the experiment, the sample is excited by two 80-fs laser pulses, $I f_1(t)$ and $I f_2(t)$, of equal intensity I and orthogonal polarizations derived from the same laser but with a variable delay τ between them, or $f_2(t) = f_1(t + \tau)$. $f(t)$ is the shape of a single pulse. If the number of electrons created in the conduction band by the pulses is not negligible, then there is a mutual saturation effect of one beam on the other while the carriers are in the photoexcited states. (Because of the vast difference in the effective density-of-state masses of the relevant electrons and holes of $\text{Al}_x\text{Ga}_{1-x}\text{As}$, the saturation effect due to the holes is not important; that is not necessarily true for other materials such as Ge, Si, or even AlAs.) As a result of the saturation effect, the total absorption of both beams will decrease when the two pulses overlap within a lifetime τ_e of each other and the measured time-averaged total transmitted power of both beams as a function of delay will show a transmission correlation peak (TCP). The change in the transmitted power of both beams through an optically thin sample due to the saturation effect is

$$\Delta T \propto \int_{-\infty}^{\infty} n_e(t) I [f_1(t) + f_2(t)] dt, \quad (1)$$

where $n_e(t)$ is the electron occupation density in the photoexcited conduction-band states:

$$n_e(t) \propto \int_{-\infty}^t I [f_1(t') + f_2(t')] \exp[-(t-t')/\tau_e] dt' = \int_0^{\infty} I [f_1(t-x) + f_2(t-x)] \exp(-x/\tau_e) dx. \quad (2)$$

The TCP corresponds to the cross terms in (1): From (1) and (2) the TCP is proportional to

$$I^2 \int_{-\infty}^{\infty} \int_0^{\infty} [f_1(t) f_2(t-x) + f_2(t) f_1(t-x)] \exp(-x/\tau_e) dx dt. \quad (3)$$

Upon changing the order of integration and substituting $f_2(t) = f_1(t + \tau)$, it can be shown that the TCP is proportional to the convolution of the intensity autocorrelation $C(\tau)$ of the pulse with a two-sided ex-

ponential decay factor: The TCP as a function of τ is proportional to

$$I^2 \int_0^\infty \left\{ \int_{-\infty}^\infty [f_1(t)f_1(t+\tau-x) + f_1(t+\tau)f_1(t-x)] dt \right\} \exp(-x/\tau_e) dx = I^2 \int_{-\infty}^\infty C(\tau-x) \exp(-|x|/\tau_e) dx. \quad (4)$$

The reason for using the two equal pulses rather than the usual pump-and-probe technique is that when the population decay is not governed by a single decay process but by a fast process (e.g., intraband relaxation) superimposed on a much slower process (e.g., band-to-band transition) as it is in the present experiment, there are real difficulties involved in using the latter technique (described in detail by Taylor, Erskine, and Tang⁶).

Because the laser has a fixed wavelength of 2.02 eV, we chose the Al concentration to be $x=0.34$ rather than 0 so that the density of states is smaller and easier to saturate. The carrier dynamics in the highly excited states of the central valley of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ are not expected to be significantly affected by this choice. A schematic of the experimental setup is shown in Fig. 1. An Ar-laser-pumped passively⁷ mode-locked rhodamine-6G-dye ring laser⁸ produces 615-nm pulses at a rate of 10^8 Hz. The autocorrelation of the pulse has a full width at half maximum of 130 fs, indicating a width of approximately 80 fs for a sech^2 pulse shape. The pulse train is divided into two beams of orthogonal polarizations by the polarizing beam splitter (PBS) which are recombined collinearly at the PBS, with the path length of one arm held fixed and that of the other dithered at ω about the position of zero path-length

difference. The combined pulse train is focused onto the sample with the spot diameter $\cong 2 \mu\text{m}$. The light transmitted through the sample is detected by a photodiode and the time-averaged power is displayed on an oscilloscope synchronized to the motion of the speaker, with the height of the TCP measured through the lock-in amplifier.

The sample is an unintentionally doped $\text{Al}_x\text{Ga}_{1-x}\text{As}-\text{GaAs}-\text{Al}_x\text{Ga}_{1-x}\text{As}$ double heterostructure grown by liquid-phase epitaxy. The GaAs substrate within a $375\text{-}\mu\text{m}$ -diam circle was etched away to allow the transmitted light through. The AlGaAs layer on the entrance side is $2.3 \mu\text{m}$ thick. The GaAs and AlGaAs layers on the exit side are $\frac{1}{3} \mu\text{m}$ and $0.1 \mu\text{m}$ thick, respectively.

Figure 2 shows examples of the measured TCP that indicate that there is saturation of the photoexcited conduction-band states. Because orthogonal polarizations are used, the corresponding relaxation time is an energy relaxation time. To determine the relaxation time for each incident power, we first digitized the data and then numerically convolved the pulse autocorrelation with an adjustable exponential decay factor. The relaxation time was determined from the best fit with the measured TCP according to Eq. (4).

Figure 4 summarizes the results on the measured

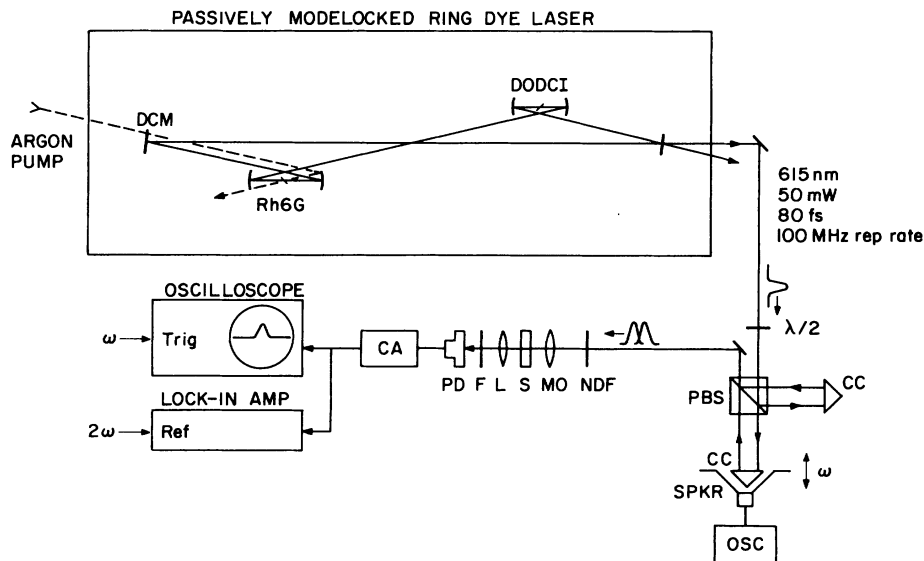


FIG. 1. Experimental setup. DCM, dichroic mirror; $\lambda/2$, half-wave plate; PBS, polarizing beam splitter; CC, corner cube prism; SPKR, speaker; OSC, oscillator; NDF, neutral-density filter; MO, $40\times$ microscope objective; S, sample; L, lens; F, monochromator or cut-off filter; PD, photodiode; CA, current amplifier. PD measures time-averaged power.

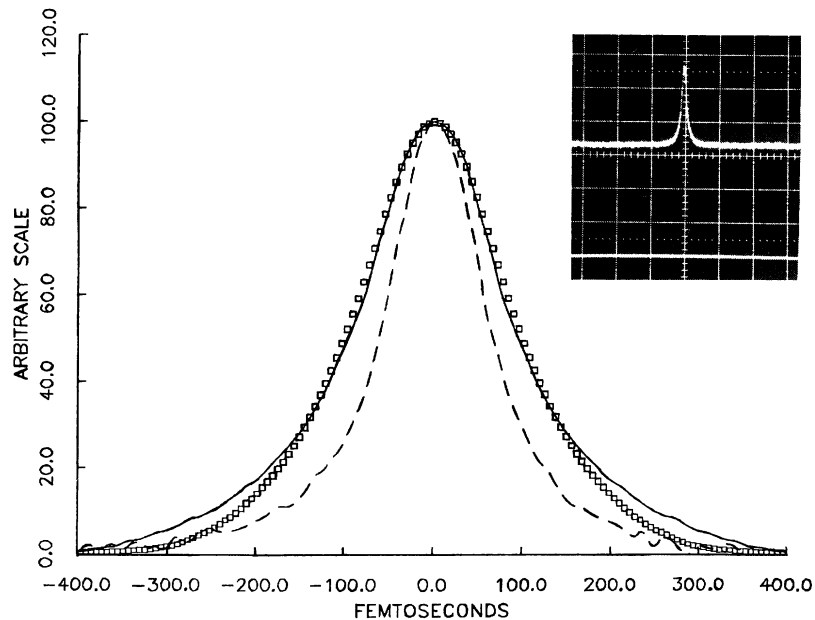


FIG. 2. Comparison of TCP at 0.63 mW (solid line) with the pulse autocorrelation (dashed line). Squares show convolution of autocorrelation peak with an exponential of decay time 55 fs. Inset shows TCP at 1.44 mW. Solid baseline indicates zero transmission. Horizontal scale is 1.1 ps/div.

relaxation time as a function of incident power using this procedure. Because of the unavoidable spatial dependence of the light intensity, the measured lifetime is clearly averaged spatially over one attenuation length.

An order of magnitude estimate of the absorbed energy per pulse needed to saturate these initial states in our sample is consistent with our interpretation that the observed effect is a saturation effect. With use of the parameters given by Monemar, Shih, and Pettit⁹ for $\text{Al}_{0.34}\text{Ga}_{0.66}\text{As}$, for our estimated saturated volume of $2.2 \times 10^{-12} \text{ cm}^3$ the total number of accessible states in the central valley is $4.2 \times 10^{-7} \Delta\nu$, where $\Delta\nu$ is determined by the bandwidth of the incident light pulse and the relaxation time of the initial states. If we take $\Delta\nu$ to be the sum of the two, or approximately $1.2 \times 10^{13} \text{ sec}^{-1}$, the total number of initially accessible states is 5×10^6 . At a photon energy of 2.02 eV, the needed absorbed energy per lifetime ($\sim 40 \text{ fs}$) to fill half of the accessible states is $8 \times 10^{-13} \text{ J}$. For a pulse length of 80 fs and a repetition rate of 10^8 s^{-1} , the combined average power for both arms is 0.3 mW. Experimentally, significant saturation (where TCP $> 10\%$ of background) is seen for powers above 0.1 mW.

This interpretation is also supported by a number of other qualitative checks. The observed power dependences of the TCP height and the

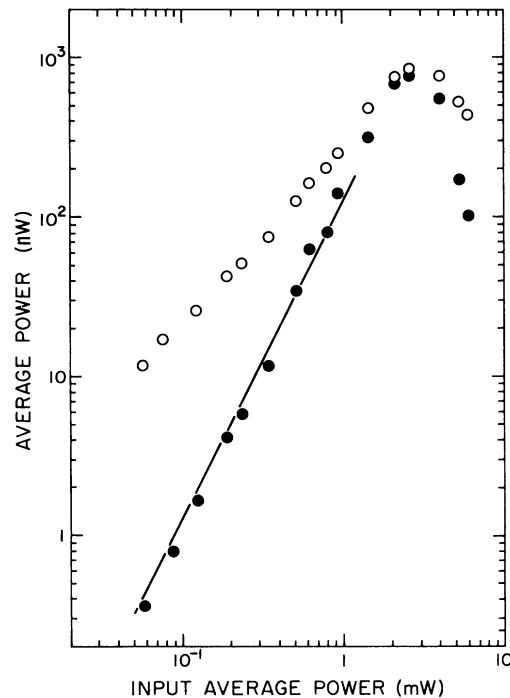


FIG. 3. Transmission signal vs total input power of both beams. Filled circles indicate the height of the TCP above background level; open circles, the background level. The solid line has a slope of 2. The strength of the transmitted signal is significantly reduced by the attenuation in the GaAs layer behind the AlGaAs layer, which changes the absolute scale but has no effect on the saturation effect.

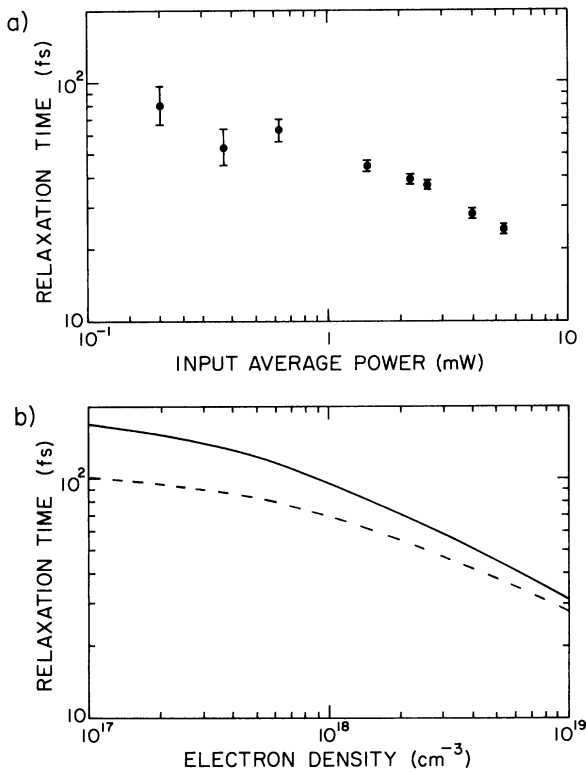


FIG. 4. (a) Measured relaxation time for $\text{Al}_{0.34}\text{Ga}_{0.66}\text{As}$ vs total input power of both beams; 1 mW corresponds to a carrier density of $7 \times 10^{18} \text{ cm}^{-3}$. (b) Calculated value for the relaxation time of GaAs without intervalley scattering (solid line) and with measured (Ref. 12) Γ to L minima intervalley scattering (dashed line).

background are consistent with the saturation effect. In the low-power range when carrier-carrier scattering is not important, the power dependence of the peak height shown in Fig. 3 is quadratic as predicted by Eq. (4). As the power increases, the carrier-carrier scattering rate increases linearly with the carrier density and the total scattering rate increases. This reduces the saturation effect and can cause the TCP to decrease with increasing incident power as indicated by the bends in the curves shown in Fig. 3. This is also consistent with the power dependence of the measured relaxation time shown in Fig. 4(a).

Since the numerical values for the parameters needed for a theoretical estimate of the relaxation rate for $\text{Al}_{0.34}\text{Ga}_{0.66}\text{As}$ are not available, it is of interest to compare our results with estimated GaAs relaxation rates. The dependence of the total relaxation time for GaAs due to carrier-carrier and OP intravalley scattering on the carrier density can be estimated on the basis of

known theories^{10,11} as shown in Fig. 4(b). Acoustical phonon scattering is neglected since it has been estimated¹¹ in GaAs to be an order of magnitude less important than OP scattering. The general shape of the calculated GaAs curve is very similar to the experimental results on $\text{Al}_{0.34}\text{Ga}_{0.66}\text{As}$ shown in Fig. 4(a). Inclusion of the measured Γ to L minima intervalley scattering¹² reduces the estimated total relaxation time for GaAs somewhat, but further inclusion of Γ to X minima scattering, of which we have no experimental or theoretical value, could reduce this even more. Judging from the density dependence of the relaxation rates, we see that the carrier-carrier scattering rate in $\text{Al}_{0.34}\text{Ga}_{0.66}\text{As}$ is also close to that indicated by the theory for GaAs. The optical-phonon scattering rate is the part independent of the carrier density.

In conclusion, we have observed directly the saturation of photoexcited Bloch states in the conduction band of a semiconductor and measured the extremely fast relaxation time from these excited states at room temperature.

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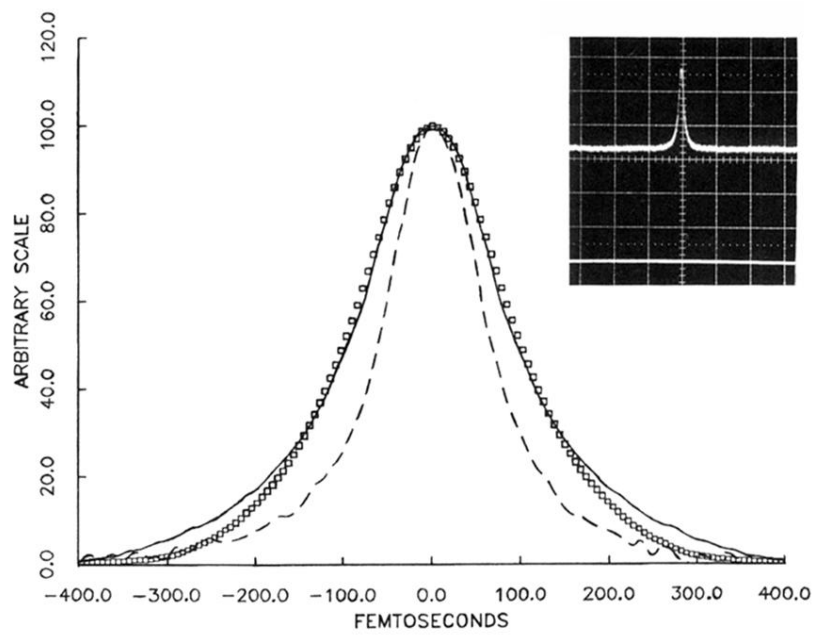


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