# Femtosecond studies of intraband relaxation in GaAs, AlGaAs, and GaAs/AlGaAs multiple quantum well structures

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Femtosecond intraband relaxation dynamics of hot carriers in highly excited states of GaAs, AlGaAs, and AlGaAs/GaAs multiple quantum well (MQW) structures are studied at room temperature using the equal-pulse correlation technique. Initial carrier lifetimes of 35, 60, and 50 fs are measured for GaAs, $Al_{0.32}Ga_{0.68}As$, and MQW structures for excitation with 2.02-eV photons at low carrier densities, and are in reasonable agreement with calculated scattering rates. The carrier-density dependence of these lifetimes is measured for densities in the range $1.5\times10^{17}$–$5\times10^{19}$ $cm^{-3}$.

The study of the dynamics of photoexcited hot carriers on femtosecond time scales in semiconductors is of fundamental importance both for the understanding of the basic physics of various scattering processes, as well as for the development of high-speed electronic devices, especially for materials such as GaAs and $Al_xGa_{1-x}As$. These carriers, generated in states far above the band edge, initially relax through processes such as intervalley and polar-optical phonon scattering, and carrier-carrier interactions to a quasi-equilibrium distribution at a carrier temperature much higher than that of the lattice. In GaAs, scattering rates for these processes are estimated to be $> 10^{12}$/s, thus indicating that a resolution of better than 100 fs is needed to observe this initial relaxation of hot carriers. Indeed, under picosecond photoexcitation, one observes only the second stage of cooling, where the hot-carrier distribution cools down to the lattice temperature through the emission of phonons. We report here the first femtosecond measurement, based on the recently developed equal-pulse correlation technique, of the initial relaxation time characterizing the isotropic depopulation of carriers from their photoexcited levels for bulk GaAs and for GaAs-AlGaAs multiple quantum well (MQW) structures. A comparison of this relaxation time between bulk GaAs and MQW structures is particularly significant in view of the recent report[1] of a significantly higher relaxation rate for the second stage of cooling in GaAs than in MQW structures following excitation by intense picosecond pulses. We also compare the experimentally determined relaxation time for GaAs and $Al_{0.32}Ga_{0.68}As$ with the theoretical values calculated from scattering rates and material parameters found in the literature.

Our measurement is based on the saturation effect in the transmission of two ~90-fs 612-nm laser pulses through a thin sample of semiconductor, using an equal-pulse correlation technique described in more detail elsewhere.[2] Briefly, in this technique the two laser pulses are orthogonally polarized, collinearly propagating, have equal powers, and a delay $\tau$ between them. The pulses are tightly focused onto the sample with an intensity such that saturable absorption takes place, and the combined time-averaged transmitted flux is measured versus $\tau$. Because of rapid depopulation of carriers from the photoexcited levels, the time-averaged transmitted flux of the pulses combined reaches a peak at $\tau = 0$ and reduces to a background value for large $\tau$. We call this peak a transmission correlation peak (TCP). It can be shown[3] that, if the depopulation of the photoexcited levels is governed by a simple relaxation time $t_r$ on the time scale of interest, and the coherent artifact is negligible,[2] then the TCP is proportional to the convolution of the laser pulse autocorrelation (AC) and a double-sided single exponential of decay time $t_r$. Thus, $t_r$ can be determined by convolving the measured AC with a double-sided single exponential and adjusting the relaxation time until a best fit is found between the convolution and the measured TCP.

This technique has been used previously to study the initial stage of relaxation of photoexcited carriers in $Al_{0.34}Ga_{0.66}As$ grown by liquid phase epitaxy.[3] For material with that fractional concentration of Al, the states photoexcited by our 2.02-eV photon are close to the band edge and thus easy to saturate. Since that time we have improved our signal processing scheme and can now obtain excellent signal-to-noise ratios for quite small TCP's (height ~0.1% of the background flux). Reference 4 describes this improved experimental setup and procedure in detail. We are now able to use this technique for the first time to study materials such as GaAs and MQW structures which produce TCP's that are an order of magnitude smaller than those observed before in AlGaAs. For comparison, we have taken additional data for an AlGaAs sample, grown by MOCVD, with our improved setup.

Three samples were measured: (1) a layer of intrinsic GaAs 0.3 $\mu$m thick cladded by 0.15-$\mu$m layers of transparent (to 612-nm light) $Al_{0.6}Ga_{0.4}As$, (2) a $\lesssim 0.3$-$\mu$m-thick unclad-ded layer of $Al_{0.32}Ga_{0.68}As$, and (3) a MQW structure having five 160-Å-thick undoped GaAs wells between 700-Å $Al_{0.6}Ga_{0.4}As$ barriers. All the samples were grown by the conventional low-pressure MOCVD process, and the substrates removed in a 375-$\mu$m circle by chemical etching.

Figure 1(a) shows a typical low-power experimental TCP scan obtained from the MQW sample. Similar scans were obtained from the GaAs and AlGaAs samples. This displays the time-averaged transmitted flux of both pulses versus delay $\tau$ between the pulses. An AC of the laser pulse, obtained using the standard method of second harmonic generation in a thin (75 $\mu$m) ADP crystal, is also shown. For comparison with the experimental MQW TCP, a convolution of the pulse AC with $t_r = 44$ fs is shown. For each sample a series of TCP's were taken for a variety of input powers,

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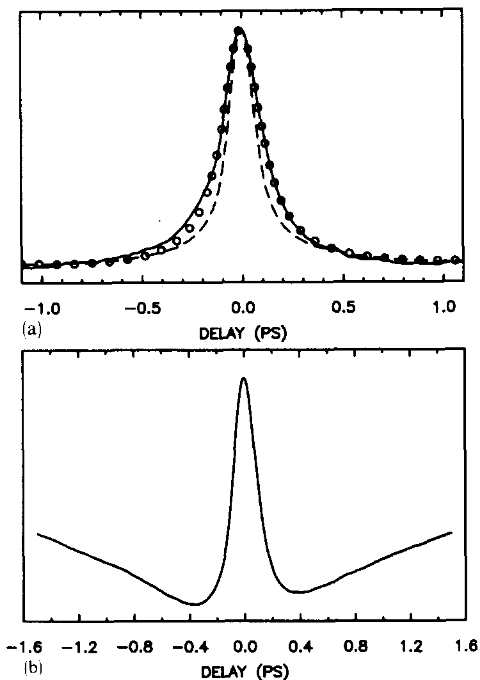


FIG. 1. (a) Solid line is a typical TCP scan of the MQW at low input power, 0.05 mW, also representative of AlGaAs and low-power scans of GaAs. The dashed line is an AC of the laser pulse, normalized to the height of the MQW TCP. The open circles correspond to the convolution of the pulse AC and the material response function $\exp(-|t/t_r|)$ for $t_r = 44$ fs. (b) A typical higher power scan of the MQW, at 0.15 mW, also representative of higher power GaAs scans. Rising wings were not seen for AlGaAs.

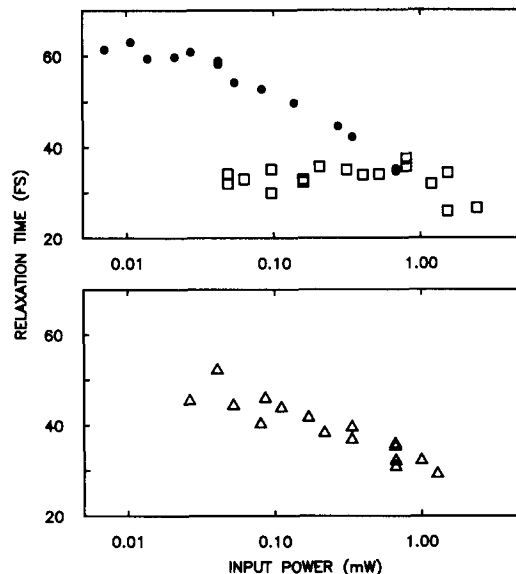


FIG. 2. Experimentally determined $t_r$ vs input laser power of both arms combined for $Al_{0.32}Ga_{0.68}As$ (circle), GaAs (square), and MQW $Al_{0.40}Ga_{0.60}As/GaAs$ structure (triangle). 1 mW corresponds to estimated average carrier densities of 1.8, 4, and $6\times10^{19}$ cm$^{-3}$, respectively, for the samples for a spot size of 1.8 $\mu$m. See text for an explanation of the set of a coherent artifact on the values of $t_r$ as shown here.

and $t_r$ was determined for each TCP scan based on the difference in width between it and the AC of the laser pulse.

In some cases our TCP shapes reflect other processes on time scales longer than the initial relaxation period. Scans for our GaAs and MQW samples (but not our AlGaAs sample) at high input powers ( $>0.1$ mW) include a rising wing, in addition to the central positive peak corresponding to the initial carrier relaxation as shown in Fig. 1(b). The characteristic time of the rising wing is $\sim 1$ ps, and the magnitude of the wing relative to the height of the positive peak increases with input power. This wing corresponds to a process occurring at high carrier densities which is clearly different from the initial relaxation from the photoexcited state. We believe this wing is caused by saturation of the transition from the split-off valence band to 0.15 eV above the bottom of the conduction band, via a band-filling process. For the GaAs and MQW data when the rising wing was discernible, mainly at high powers, the background "valley" formed by the rising wings was subtracted away leaving a positive peak similar to that in Fig. 1(a).

In Fig. 2 we present our experimental results of relaxation time $t_r$ versus input power for the three materials at room temperature. At low powers, where carrier-carrier scattering is not expected to be important, $t_r$ is determined by the intervalley (IV) and polar-optical (PO) phonon scattering rates. Thus, these data indicate these 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 IV and PO 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. Because of the unavoidable spatial dependence of the intensity through the sample, the measured lifetime corresponds to that with the photogenerated carrier density averaged over an attenuation length.

Following intense femtosecond excitation, we see only a slight increase in the relaxation times in Fig. 2 between the MQW structure and GaAs. In contrast, following intense picosecond excitation at 77 K (carrier density $\gg 10^{17}$ cm$^{-3}$), a significant difference in the rates corresponding to the second stage of cooling was reported[1] between MQW and bulk GaAs; it was hypothesized that this difference was due to the trapping of phonons generated in the QW region. Since the femtosecond relaxation we observe here has to do with the initial emission of phonons, and the electronic densities of states in bulk GaAs and the MQW structure are comparable, the initial femtosecond relaxation rates in the two cases are expected to be comparable.

In our experimental configuration, there should be no significant coherent artifact[5] when the momentum reorientation time within the optically coupled states is faster than the pulse width and the energy relaxation time out of those states or when the photoexcited electron distribution is isotropic. Comparison of preliminary measured and calculated ratios of the TCP heights under parallel and perpendicular polarization configuration indicate that this may be a 20% coherent artifact contribution to our observed TCP. The values for $t_r$ are plotted in Fig. 2 assuming no coherent contribution. A coherent artifact contribution of 20% would increase the values for $t_r$ by about 30%.

We have calculated the scattering rates expected to be important in the low carrier density limit from expressions[6] and material parameters[7] found in the literature. These rates are shown in Table I for electrons and holes in the two materials. While the expression for PO scattering uses relatively



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TABLE I. Calculated intervalley ($R_{iv}$) and polar optical ($R_{+,-po}$) scattering rates for carriers generated by a 2.02-eV transition from the heavy-hole valence band at 300 K based on expressions in Ref. 6, a deformation potential[8] $D_{\Gamma X} = D_{\Gamma L} = 10^9$ eV/cm, and commonly estimated material parameters.[7] $R_{iv}$ is the $\Gamma$-$L$ intervalley rate alone, with parentheses enclosing values when the maximum expected value for the $\Gamma$-$X$ intervalley rate is added to the $\Gamma$-$L$ intervalley rates. $R_{+po}$, $R_{-po}$ are rates for absorption and emission, respectively, of a PO phonon.

| Material | GaAs | | $Al_{0.32}Ga_{0.68}As$ | |
|---|---|---|---|---|
| carrier | electron | hole | electron | hole |
| $R_{iv}$ ($10^{12}$/s) | 25(35) | 0 | 15(25) | 0 |
| $R_{+po}$ ($10^{12}$/s) | 1.7 | 5.5 | 2.2 | 6.8 |
| $R_{-po}$ ($10^{12}$/s) | 6.4 | 18.5 | 7.9 | 0 |

well-known material parameters, the expression for IV scattering between valleys $i$ and $j$ requires the deformation potential $D_{ij}$, the value of which is poorly known. We use the values determined empirically by Littlejohn *et al.*[8] in their Monte Carlo simulation of high-field transport in GaAs. The values for AlGaAs are assumed to be the same. Additionally, the location of the $X$-valley minimum in energy is not known with certainty in either material; it is possible that the $\Gamma$-$X$ intervalley rate is much smaller than the $\Gamma$-$L$ rate. We have calculated the rates for the cases when there is only $\Gamma$-$L$ intervalley scattering, and when the maximum expected contribution from $\Gamma$-$X$ scattering is included.

To compare measured $t_r$ to the calculated scattering rates, we note that $t_r$ is the relaxation time describing the decay of the difference between electron populations in the conduction and valence-band states in $k$-space isoenergetic to the initially excited states. At low carrier density ($< 1.6 \times 10^{18}$ cm$^{-3}$) we have measured $t_r$ to be 35 fs in GaAs and 60 fs in AlGaAs. By comparison, based upon the scattering rates given in Table I (for IV scattering to the $L$ valley only), the initially excited electron and hole populations should decay with approximate time constants of 30 and 60 fs, respectively, in GaAs and 40 and 150 fs, respectively, in AlGaAs. Since the quasi-equilibrium distribution reached after the first stage of cooling does not significantly fill the optically coupled region in either the valence band or the conduction band, $t_r$ characterizes the decay of the sum of the electron and hole populations. Because the decay times for the electron and hole populations are different, their sum cannot be characterized by a single exponential decay over a long period of time. Our calculated value for $t_r$ is determined by the decay of the sum of the electron and hole populations to its $1/e$ height. From our calculated electron and hole relaxation times we estimate $t_r$ to be 40 fs for GaAs and 80 fs for AlGaAs, in qualitative agreement with our measured values.

In conclusion, we measured the initial relaxation time characterizing the isotropic depopulation of carriers from their initially photoexcited levels as a function of carrier density $n$ in the range $n = 1.5 \times 10^{17}$–$5 \times 10^{19}$ cm$^{-3}$. At low carrier densities, relaxation times of about 35, 60, and 50 fs were observed for GaAs, $Al_{0.32}Ga_{0.68}As$, and MQW structures, respectively. Inclusion of a possible coherent artifact contribution would increase these values by about 30%. These measured relaxation times are consistent with the calculated intervalley and polar-optical phonon scattering rates using a deformation potential on the order of $10^9$ eV/cm.

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