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## FEMTOSECOND TIME-RESOLVED REFLECTIVITY OF Ge

K.D. MYERS, S. ZOLLNER, R. LANGE, K.G. JENSEN,<sup>a</sup> J.M. DOLAN  
*Ames Laboratory and Department of Physics and Astronomy,  
Iowa State University, Ames, IA 50011, USA*

D.W. BAILEY<sup>b</sup>  
*Department of Electrical and Computer Engineering,  
University of South Carolina, Columbia, SC 29208, USA*

C.J. STANTON  
*Department of Physics, University of Florida, Gainesville, FL 32611, USA*

We have measured the transient reflectivity changes of bulk Ge after excitation with 140 fs laser pulses at 1.5 eV. The electron and hole carrier dynamics are calculated using an ensemble Monte Carlo method. The observed reflectivity changes are due to three mechanisms: Diffusion, band gap renormalization, and carrier dynamics, particularly scattering of light holes to the heavy hole band via optical phonons.

### 1 Introduction and Summary

For many years, femtosecond laser spectroscopy has focussed on III-V semiconductors and only a few picosecond studies of Ge<sup>1-3</sup> were conducted. This has changed recently,<sup>4-7</sup> partly due to the femtosecond titanium-sapphire laser and a revived interest in Ge and Si<sub>1-x</sub>Ge<sub>x</sub> alloys. Previous studies using photoluminescence,<sup>6</sup> transmission,<sup>7</sup> and four-wave mixing<sup>1,4</sup> have focussed on carrier cooling,<sup>6</sup> electron dynamics such as intervalley scattering,<sup>4,6,7</sup> band gap renormalization,<sup>7</sup> and diffusion.<sup>1,2</sup> In this work, we concentrate on the hole dynamics, particularly the scattering of light holes to the heavy hole band. We find that an optical deformation potential constant of  $DK=10$  eV/Å overestimates the light-hole to heavy-hole scattering rate by about a factor of 2.

### 2 Experimental Procedure and Results

Time-resolved photoinduced reflectivity changes ( $\Delta R/R$ ) of bulk intrinsic Ge were measured at 300 K in the usual slow-scan pump-probe geometry using 140 fs pulses at 1.5 eV from a titanium-sapphire laser, see the thick solid line in Fig. 1 (a). At small negative time delays  $\tau$ , there is an increase in  $\Delta R/R$  followed by a sharp decrease which peaks near  $\tau=0.3$  ps. The reflectivity then

<sup>a</sup>Now at Quantum Design Inc., San Diego, CA 92121.

<sup>b</sup>Now at Digital Equipment Corporation, 77 Reed Rd., Hudson, MA 01749.

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recovers within about 6 ps. The surface carrier density (calculated from the energy and spot size of the laser) is about  $4 \times 10^{18} \text{ cm}^{-3}$ .

### 3 Discussion

Unlike transmission, which only measures absorption changes at the probe wavelength, the reflectivity is related to the Kramers-Kronig transform of the absorption and therefore depends on a variety of factors. This has advantages as well as disadvantages. The reflectivity changes  $\Delta R$  measured here are mostly due to the photoexcited carriers (created by the pump pulse) performing plasma oscillations at the probe laser frequency  $\omega_L$ , thus screening the electric field of the probe laser. Within the Drude model,<sup>5</sup> the free-carrier contributions to the dielectric function  $\epsilon$  can be calculated from the plasma frequency  $\omega_p$ , which depends on the sum of the ratios of the carrier densities  $n$  to the effective masses  $m$  in all electron valleys (e) and hole bands (h)

$$\Delta\epsilon(\omega_L, \tau) = -\epsilon_s \left( \frac{\omega_p(\tau)}{\omega_L} \right)^2, \quad \omega_p^2(\tau) = \frac{e^2}{\epsilon_0 \epsilon_s} \left[ \sum_e \frac{n_e(\tau)}{m_e} + \sum_h \frac{n_h(\tau)}{m_h} \right], \quad (1)$$

where  $\tau$  is the delay time between the pump and the probe pulse. The reflectivity change  $\Delta R$  is then calculated from  $\Delta\epsilon$ .

We use the results of an ensemble Monte Carlo simulation<sup>8</sup> to model the experimental data. The electron and hole concentrations as a function of  $\tau$  are shown in Figs. 5 and 7 of Ref. 8: Electrons are created in the  $\Gamma$ -valley. They initially scatter to the satellite valleys (predominantly the  $X$ -valley because of its higher density of states) within 100 fs. After about 4 ps, most electrons have relaxed to the bottom of the  $X$ -valley and then have scattered to the  $L$ -valley, which is the global minimum of the conduction band in Ge. The Monte Carlo simulation shows that 25% of the holes are created in the light hole band. They scatter to the heavy-hole band with a time constant of about 1 ps, which can easily be resolved with the 140 fs laser pulse. Additionally, this time constant is much longer than the lifetime of  $\Gamma$ -electrons.<sup>4,6,7</sup> These simulations were performed for conditions similar to the experiment (1.5 eV pulses with a half-width of 100 fs, surface carrier density  $10^{18} \text{ cm}^{-3}$ ), but diffusion and band gap renormalization were not included. We can scale the Monte Carlo results by a factor of 4 to obtain results for  $n=4 \times 10^{18} \text{ cm}^{-3}$ , since most scattering rates are approximately linear in density, at least for a small range of densities. (This has been confirmed by performing simulations at different densities.) Because of the large density of states at  $X$  and  $L$  in Ge, Pauli blocking does not affect the electrons as much as in the  $\Gamma$ -valley in GaAs for densities near  $10^{18} \text{ cm}^{-3}$ .

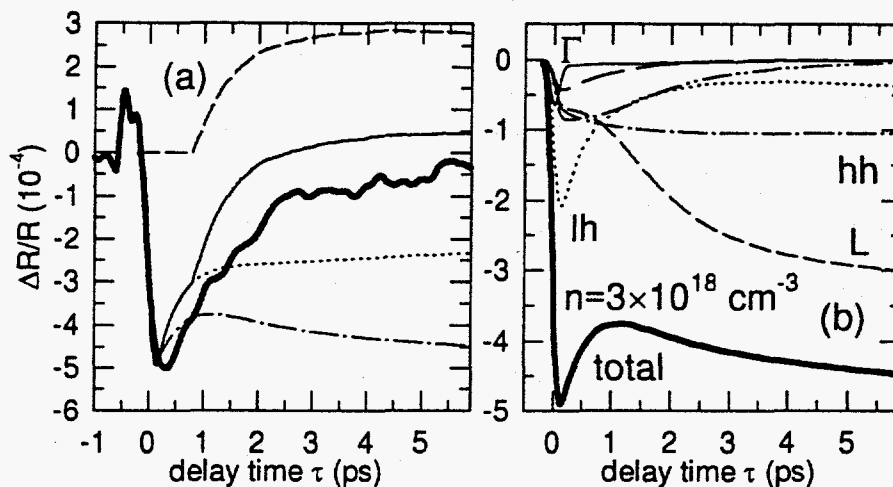


Figure 1: (a) Measured reflectivity change as a function of delay time  $\tau$  between the pump and the probe pulse (thick solid line). Calculated reflectivity contributions: Monte-Carlo (MC) only (dot-dashed line, same as the thick solid line in (b)), band gap renormalization (BGR) only (dashed), MC plus diffusion (dotted), MC plus diffusion plus BGR (thin solid line). (b) Nonequilibrium Drude reflectivity of photoexcited carriers calculated from the carrier distribution found in the Monte-Carlo simulation: Heavy (dot-dashed), light (dotted), and split-off holes (long-dashed). L- (dashed), X- (double dot-dashed), and  $\Gamma$ -valley (thin solid line) electrons.  $\Delta R/R$  from carriers in all six bands is given by the thick solid line. Diffusion and BGR are not considered.

Figure 1 (b) shows the contributions to the Drude reflectivity from the electrons and holes in different bands (calculated from the Monte Carlo densities). It can clearly be seen that the peak at 0.2 ps is due to the occupation of the light hole band. The peak decreases, as the light holes scatter to the heavy hole band. The slow increase after 1 ps is due to the scattering of electrons from the X-valleys to the L-valleys. The total calculated transient Drude reflectivity (thick solid line in Fig. 1 (b)) is compared with the experimental data in Fig. 1 (a) (dot-dashed line). The agreement is not good, since we have not yet corrected the Monte-Carlo densities to account for diffusion and band gap renormalization.

Corrections due to diffusion were calculated by solving the diffusion equation with a time-dependent diffusivity. The diffusivity as a function of carrier temperature is given by Smirl.<sup>1</sup> The average carrier temperature can be obtained from the Monte Carlo simulation. The reflectivity calculated from the Monte-Carlo densities, corrected for diffusion, are given by the dotted line in Fig. 1 (a).

Finally, we consider reflectivity changes due to band gap renormalization (BGR) using Zimmermann's model.<sup>9</sup> The carrier temperature was again taken from the Monte Carlo simulation. We also have to consider the fact that only about 80% of the carriers recombine before the arrival of the next laser pulse, resulting in carrier accumulation. (This number can be obtained by solving a steady-state diffusion equation containing an Auger recombination term.) The resulting BGR corrections are given by the dashed line in Fig. 1 (a). The thin solid line gives the reflectivity calculated with our final model, including carrier dynamics, diffusion, band gap recombination, and carrier accumulation.

We stress that the comparison between theory and experiment in Fig. 1 (a) contains only one fit parameter: The spot size of the pump laser. The agreement can be improved by fine-tuning the Auger recombination rate (at late times) and the optical deformation potential for holes. The results suggest that the optical deformation potential constant of  $DK=10$  eV/Å overestimates the light-hole to heavy-hole scattering by a about a factor of two. The positive peak at negative time delays was also observed in Ref. 7 and attributed to BGR effects. However, our BGR model was unable to account for this peak.

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