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FREE CARRIER AND LATTICE-HEATING-INDUCED CHANGES TO THE REFLECTIVITY OF EPITAXIAL GeSi ALLOYS FOLLOWING PICOSECOND PULSE EXCITATION

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Transient reflectivity characteristics for epitaxial $Ge_{1-x}Si_x$ (x=0, 0.05, 0.1, 0.25) alloys have been measured at $\lambda = 0.575\mu m$, using 2ps pump and probe pulses with photogenerated carrier densities up to 10^{19} cm^{-3}. For all x we are able to separately determine negative (free carrier) and positive (lattice heating) contributions to the reflectivity change; the relative contributions of the latter are found to increase with increasing x. The evolution of the free carrier part is dominated by carrier-temperature-dependent diffusion.

Over the past two decades there has been considerable research on transient carrier kinetics and optical properties of the Group IV semiconductors[1-4] Ge and Si following picosecond excitation, and these materials have often served as the testbed for studies of ultrafast phenomena in semiconductors. One effect which has been the subject of numerous investigations in these and polar semiconductors is the energy relaxation of photo-excited carriers and the associated generation of nonequilibrium optical phonons.[5] Although several studies have been performed to investigate either carrier or phonon dynamics, recently[6] we have been able to correlate the hot-carrier and the hot phonon dynamics in intrinsic Ge using picosecond, time-resolved reflectivity and Raman scattering at $\lambda = 0.575$ μm. In the interpretation of those experiments it became necessary to consider the relative roles of photo-excited carriers versus lattice heating in effecting a change in the optical properties, a nonambiguous task for experiments performed at a single wavelength. There was also a question of whether a non-equilibrium optical phonon population can bring about the same change in dielectric constant as the lattice thermal equilibrium state (at elevated temperture) which follows after tens of picoseconds if not longer. Although suggestions concerning these questions have been made for silicon-on-saphire[7], in that case the deconvolution of lattice heating effects from carrier induced effects was complicated by the effective Fabry-Perot geometry of the specimen.

Information about the relative roles and time-dependence of free carrier and lattice heating induced contributions to the refractive index of semiconductors is also important because of the strong interest in applications of light-by-light switching.[8] In an attempt to answer these questions we have conducted picosecond, time-resolved reflectivity experiments on a number of Ge-Si alloys for which the band gap and temperature dependence of the refractive index varies strongly with silicon fraction. We show that for the Ge-Si alloys there is clear evidence that lattice heating can induce substantial changes in the optical properties on a picosecond time scale. In addition, the results presented here provide some of the first evidence of transient carrier processes in Ge-Si alloys. We note that experiments such as these, in which the probe-frequency is constant and the material band gap is varied, can provide analogous information to that given by varying the probe-frequency in a single material.

The optical source for the experiments was a mode-locked and cavity-dumpled dye laser which produced pulses with $\lambda = 0.575$ μm and a width of 2.5 psec at a repetition rate of 3.8 MHz. The pump-probe experimental technique employed to measured the time-resolved reflectivity change has been reported elsewhere.[6] $Ge_{1-x}Si_x$ single crystals with $x \simeq 0.05$, 0.1 and 0.25 were grown on (100) Ge substrates using molecular beam epitaxy techniques to thicknesses of greater than 0.2 μm.[9] Table I gives a summary of the linearly interpolated band gap energies, E_g densities, ρ, specific heats, C_L, absorption coefficients, α, ambient diffusion coefficients, D^o, (all from the literature), and the refractive index temperature coefficients, $\frac{dR}{dT_L}$ deduced from our experiments. Since the epitaxial layers are partially strained, the linearly interpolated material parameters are only approximate, but the errors due to these discrepancies will be small given the other assumptions in the model. All experiments were conducted at room temperature. For the average irradiance we estimate that the steady-state increase in lattice temperature is no more than 50K while the surface temperature increase following a particular pulse is on the order of 5K.

Figure 1 shows the time-resolved reflectivity in the different alloys for a pulse fluence of $1 mJ/cm^2$. For comparison, results are also shown from previous work on pure germanium. All data sets show a rapid drop in reflectivity characteristic of the rise time of the pulse. This is followed by a rise, initially rapid and then more gradual with increasing delay. With increasing Si fraction one observes that the minimum reflectivity (at $t \simeq 1$ psec) becomes less negative while the long term reflectivity increases, and actually becomes positive for x=0.25. It is noteworthy, that to within experimental error, all the data sets for $t > 1$ psec

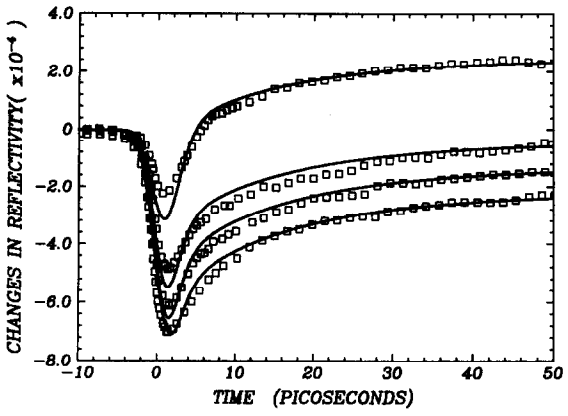

Fig 1. Time resolved reflectivity data for epitaxial $Ge_{1-x}Si_x$ alloys (for x=0.25, 0.10, 0.04, 0, top to bottom) following excitation by 2psec, λ=0.575μm pulses with irradiance of 1mJ/cm². Vertical error bars are approximately twice the size of the squares. Curves are theoretical fits with details discussed in text.

can be made to fall on top of each other if one fixes the minima to fall on top of each other. That is, the *change* in reflectivity, after the minimum is reached following the pulse, is virtually the same in each case.

In order to interpret the data we have extended the model[6] used to explain the transient reflectivity characteristics of pure Ge. There, we were able to achieve reasonable agreement between the data and a kinetic model for carrier generation and transport based on the Boltzmann equation, with phonon generation and lattice heating determined by a microscopic model of carrier- phonon interactions. This model is considered here for parameters relevant to the alloy specimens (see Table I). The reflectivity characteristics were modelled by carrier density and lattice temperature induced changes to the dielectric constant (ε), where as an approximation we assumed that the non-equilibrium phonons generated during the pulse thermalize with the other phonon modes instantly. This is equivalent to saying that the variation of dielectric constant depends only on lattice energy content and not a thermal equilibrium state. It follows that

$$\varepsilon = \varepsilon_0 + \Delta\varepsilon \qquad (1)$$

where ε_0 is the quiescent dielectric constant for the particular alloy and where the changes in dielectric constant due to carriers of density N and an increase in lattice temperature (T_L) are given by

$$\Delta\varepsilon = \Delta\varepsilon(N) + \Delta\varepsilon(T_L). \qquad (2)$$

Expressions for the free carrier contributions to Δε are well known[10] and can be approximated by Lorentz and Drude contributions. At a fixed wavelength of the probe pulse the free carrier and inter-band parts are both proportional to the carrier density, N, and hence we can take

$$\Delta\varepsilon = -AN, \qquad (3)$$

where A is a (sample dependent) constant. The change in reflectivity can then be related to the dielectric constant through the usual Fresnel relations so that we have

$$\Delta R = \Delta R(N) + \frac{dR}{dT_L}(T_L - 350K) \qquad (4)$$

where the first term on the right hand side includes contributions by carriers and the second is related to induced lattice heating. The model treats the density and temperature parameters as functions of distance from the surface and time, and hence the inhomogeneous nature of the plasma is taken into account. There are thus two variable (sample specific) parameters in the model, A and $\frac{dR}{dT_L}$

A key feature in understanding the data for pure Ge is that the decay characteristics of the reflectivity are governed by a carrier-temperature-induced diffusion coefficient which decreases as a function of time.[6] This can be related to the carrier temperature dependence of the diffusion coefficient following carrier generation and heating. As noted above, the recovery dynamics of the reflectivity after it reaches its minimum value, is essentially identical for all of the alloys and pure Ge. This implies that the peak injected carrier density and the diffusion dynamics of the optically-pumped plasmas are very similar in all alloys. The most significant difference in the reflectivity results is the *uniform* increase in the reflectivity during and following pulsed excitation. Because this effect occurs for short as well as long times it cannot be related to carrier effects, such as carrier temperature dependences of the inter-band contribution to the dielectric constant[10-12] since the carrier temperature approaches that of the lattice within the duration of the pulse (see Fig. 2). We therefore attribute the rise in the minimum reflectivity and the long term increase in ΔR with increasing x to effects of lattice heating. This

TABLE I
PROPERTIES OF $Ge_{1-x}Si_x$

x	E_g (eV)[19]	$\frac{dR}{dT_L}$ ($\times10^{-5}$), all ±50%	ρ(g/cm³)[14]	D_e^o[15] (cm²/s)	D_h^o[15] (cm²/s)	C_L[16,17] (J/cm³/K)	α (cm⁻¹)[18]
0	0.66	<1	5.32	103	54	1.70	3.0×10^5
0.05	.68	1	5.17	100	52	1.71	2.8×10^5
0.1	.70	2.5	5.02	96	50	1.73	2.7×10^5
0.25	.77	6.0	4.57	86	44	1.77	2.3×10^5

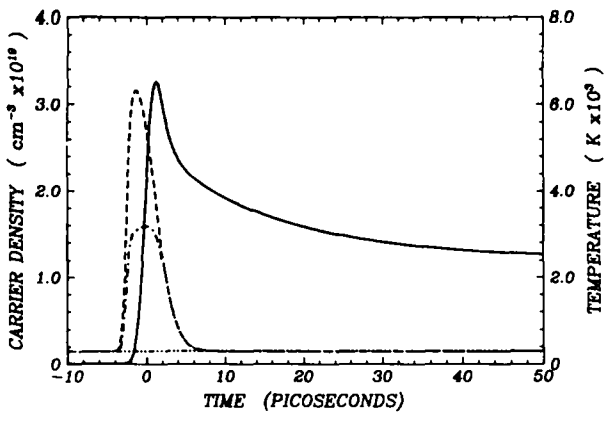

Fig 2 Model predictions for surface carrier density (solid), electron (dashed) and hole (dash-dotted) temperatures, and "lattice temperature" (dash-dot-dotted) under excitation conditions corresponding to Fig. 1. The results are shown for the x=0.25 alloy.

increased sensitivity of the alloys to lattice heating is largely due to a temperature coefficient $\frac{dR}{dT_L}$, which increases with increasing silicon content (see Table 1), since the model results show only a weak dependence of the induced lattice temperature change on the alloy composition.

The similarity of the carrier-induced changes in all samples is not surprising since the relevant material parameters (Table I) are not drastically different (typically a 10-20% variation), and the change in absorption coefficient is largely compensated by a reduced diffusion rate (both the ambient diffusion coefficients D_e^o and D_h^o, and the excess carrier temperatures, $\alpha(h\nu-E_g)$, are less in the alloys). As a trend, the sign of, and the increasing magnitude of the temperature coefficient $\frac{dR}{dT_L}$ with increasing x is not inconsistent with the reflectivity spectra from bulk, unstrained alloys in the literature.[13] However, we are unaware of any directly relevant data in the literature with which we might compare the absolute values.

In the numerical simulations, all of the energy transferred to the lattice from the carriers is assumed to instantaneously increase the lattice temperature. Energy transfer from the carriers to the optical phonons occurs within 2 psec in Ge. However, from the Raman scattering results on germanium it is known that it takes on the order of 4 psec for the energy transferred to the zone-centre optical phonons to *start* equilibrating with the rest of the lattice at 300K. The corresponding time for other phonon modes that directly couple to the carrier system is unknown. The fits to the transient reflectivity data for the alloys obtained assuming instantaneous equilibration are reasonable, but there is a systematic discrepancy for delays up to ~15 ps. This discrepancy is such that it would be *worse* if we attempted to delay the (positive) contribution of the increased lattice energy to the reflectivity. To explain the discrepancy in terms of the lattice contribution to ΔR, it would be necessary to postulate that the effect of a given energy-density resident in the lattice system is *larger*, and of the same sign, when it is *athermally* distributed in high-energy modes, than when it is equilibrated. We can not completely rule out this possibility, but it should be noted that the discrepancy could also be due to a systematic inaccuracy in the model's treatment of carrier-temperature dependent diffusion. The model and quality of fit in Ge are certainly good enough to demonstrate the inadequacy of assuming a constant diffusion coefficient,[6] but given the simplifying approximations, they may not be good enough to claim *complete* understanding of the carrier contributions to ΔR, especially in the alloys.

In conclusion we have observed transient reflectivity characteristics of Ge-Si alloys following picosecond pulse excitation. In a number of alloys, the transient effects are similar and consistent with hot carrier diffusion followed by normal diffusion. With increasing Si content we see that lattice heating becomes increasingly important in making contributions to the change in refractive index.

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