Ultrafast initial relaxation of hot electrons and holes in tetrahedral semiconductors via deformation potential interaction: Theory and experiment

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The broadenings of the E_1 and $E_1+\Delta_1$ interband critical points can be understood as lifetime effects due to the ultrafast relaxation of the photoexcited hot holes. The contributions to these broadenings arising from the electrons in the conduction band are small, as intervalley scattering times are rather long. We have measured such broadenings in Si, Ge, α-tin, AlAs, AlSb, GaP, GaAs, GaSb, InP, InAs, and InSb with spectroscopic ellipsometry and compare them with calculations based on the deformation potential-type electron-phonon interaction in the rigid pseudo-ion approximation.

The absorption of tetrahedral semiconductors is moderate ($\leqslant 5\times 10^4$ cm^{-1}) near the lowest direct gap E_0, but rises sharply at the E_1 and $E_1+\Delta_1$ gaps with energies ranging from 2.2 eV in InSb to 4.0 eV in AlAs.[1] These features in the absorption spectra are caused by interband transitions from the $\Lambda^v_{4,5}$ and Λ^v_6 valence bands to the Λ^c_6 conduction band[2] and can be studied using ellipsometry[1] or other reflection modulation techniques.[3,4]

Because of these optical properties, visible or near ultraviolet light photoexcites hot holes with very large wave vectors and energies up to 1 eV or more in the upper valence bands. These holes will scatter within femtoseconds out of their initial states by emitting or absorbing phonons via the deformation-potential interaction and relax towards the valence band top. This interaction can be treated within the rigid-pseudoion method, based on lattice dynamical models for the phonons and an empirical local pseudopotential band structure.[5] The photoexcitation also creates electrons, of course, but these relax more slowly, since the density of final states in the conduction band near Γ is small.[6] The purpose of this work is to show that the widely studied effect of intervalley scattering in the conduction band[6] has an analog in the valence band, which can be studied experimentally using spectroscopic ellipsometry and described theoretically with the rigid-pseudoion method.[6]

The initial relaxation of a hole with wave vector k and band index n photoexcited in one of those processes, i.e., the emission (or absorption) of the first phonon, takes about 5 fs (at 300 K) and can hardly be studied in real time. However, it causes a temperature-dependent Lorentzian broadening $\Gamma_{kn}(T)$ related to the scattering time $\tau_{kn}(T)$ by the relation[5]

$$\Gamma_{kn}(T)=\hbar/2\tau_{kn}(T)=329\ \text{meV fs}/\tau_{kn}(T), \tag{1}$$

which can be measured with cw techniques like ellipsometry. $\Gamma_{kn}(T)$ can be expressed in terms of a temperature-independent electron-phonon spectral function $g^2B(\mathbf{k},n;\Omega)$ as[5]

$$\Gamma_{kn}(T)=\int_0^\infty d\Omega\, g^2B(\mathbf{k},n;\Omega)\left(N_\Omega(T)+\frac{1}{2}\right), \tag{2}$$

where Ω is the energy of the emitted or absorbed phonon and $N_\Omega(T)$ the Bose–Einstein occupation factor.

Typical spectral functions for electrons and holes in GaSb with wave vectors in Λ direction, calculated using the pseudopotential form factors of Ref. 7 (without inclusion of spin-orbit interaction) and the lattice dynamical model of Ref. 8, are shown in Figs. 1(a) and 1(b). We integrate g^2B according to Eq. (2) to obtain the temperature-dependent broadenings, as shown in Fig. 1(c). The relaxation times can be obtained from Eq. (1) using the corresponding values of the broadenings of Fig. 1(c). These times are rather long for electrons, but much shorter for the holes, because the density of final states in the corresponding valence bands is much larger than that for the conduction band.

We average the calculated broadenings along the Λ direction between the points $L/4$ and L and sum the contributions of electrons (dotted lines in Fig. 2) and holes (dashed lines) as representing the total calculated broadenings (solid lines) of the E_1 gap and compare them with the ellipsometric data (symbols). We stress that the broadenings are dominated by the holes (except for Si).

The experimental broadenings can be obtained from the second or third derivatives of the ellipsometric spectra with a fit to analytic line shapes.[4] It is difficult, however, to obtain unambiguous results, as it is not clear to what extent excitonic effects (saddle point exciton) should be taken into account.[9] Results for GaSb, obtained from fits with both 2-D and excitonic line shapes, are presented in Fig. 2; they compare well with the calculated broadenings. Similar agreement between theory and experiment has been achieved for Ge, Si, and GaAs (published elsewhere[5]), as

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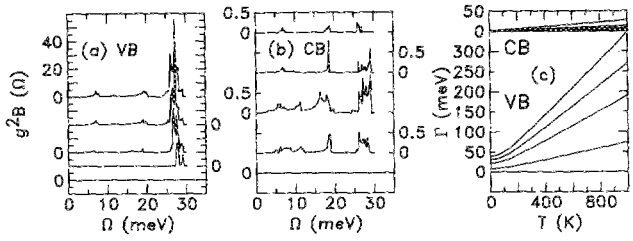

FIG. 1. Electron-phonon spectral functions for (a) holes (VB) and (b) electrons (CB) in GaSb along the Λ direction with wave vectors $k = \Gamma$, $L/4$, $L/2$, $3L/4$, and L (bottom to top) together with (c) the temperature-dependent broadenings for these states obtained from the spectral functions (without spin-orbit interaction).

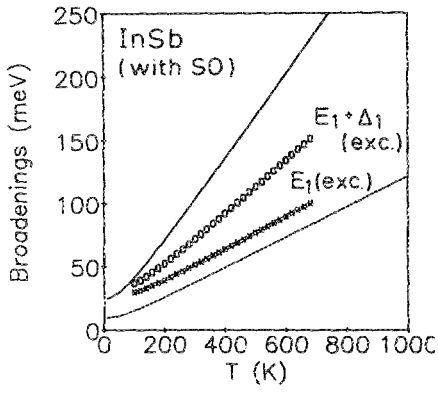

FIG. 3. As Fig. 2, but with the spin-orbit interaction included for InSb. The dashed line shows the calculated broadening for E_1, the solid line for $E_1 + \Delta_1$.

well as for α-tin,[10] AlAs,[11] AlSb,[12] InP,[13] InAs, and InSb[14] (see Fig. 2). In GaP, the spin-orbit splitting Δ_1 is too small to be resolved in ellipsometry. Therefore, the two structures E_1 and $E_1 + \Delta_1$ had to be fitted as one critical point, thus overestimating the broadening; see Fig. 2.

On the other hand, in InSb[14] Δ_1 is large ($\sim$800 meV). This splits the doubly degenerate bands Λ_3^v and reduces the density of final states for initial states in the highest valence band $\Lambda_{4,5}^v$ by a significant amount. Therefore, a calculation without spin-orbit splitting overestimates the broadenings for E_1. If spin-orbit splitting is taken into account, the broadenings of the E_1 gap will decrease and those of the $E_1 + \Delta_1$ gap increase. We have included these spin-orbit splittings using the method of Ref. 15. The resulting broadenings are shown in Fig. 3 (dashed line: E_1, solid line: $E_1 + \Delta_1$) and are in reasonable agreement with the experiment, especially for E_1. The fact that $E_1 + \Delta_1$ shows a slower increase in Γ with T than calculated may be a quirk of the fitting procedure[9] due to the fact that $E_1 + \Delta_1$ is weaker than E_1.

The relaxation times discussed so far are phase relaxation times. It is also possible to calculate the energy relaxation rates, taken as the difference between the phase relaxation times for absorption and emission multiplied with the phonon energy:

$$\frac{\partial E}{\partial t} = -\frac{1}{\hbar}\int_0^\infty \Omega d\Omega\, g^2 B(\mathbf{k},n,\Omega). \tag{3}$$

These relaxation rates are temperature independent and given in Table I for some materials.

In summary, we have calculated the ultrafast initial relaxation times of electrons and holes with wave vectors in the Λ direction. When the lifetime broadenings of electrons and holes are added up and averaged over some points along the Λ direction, we obtain reasonable agreement with experimental broadenings of the E_1 and $E_1 + \Delta_1$ critical points observed with temperature-dependent spectroellipsometry. For heavier compounds (InSb), the spin-

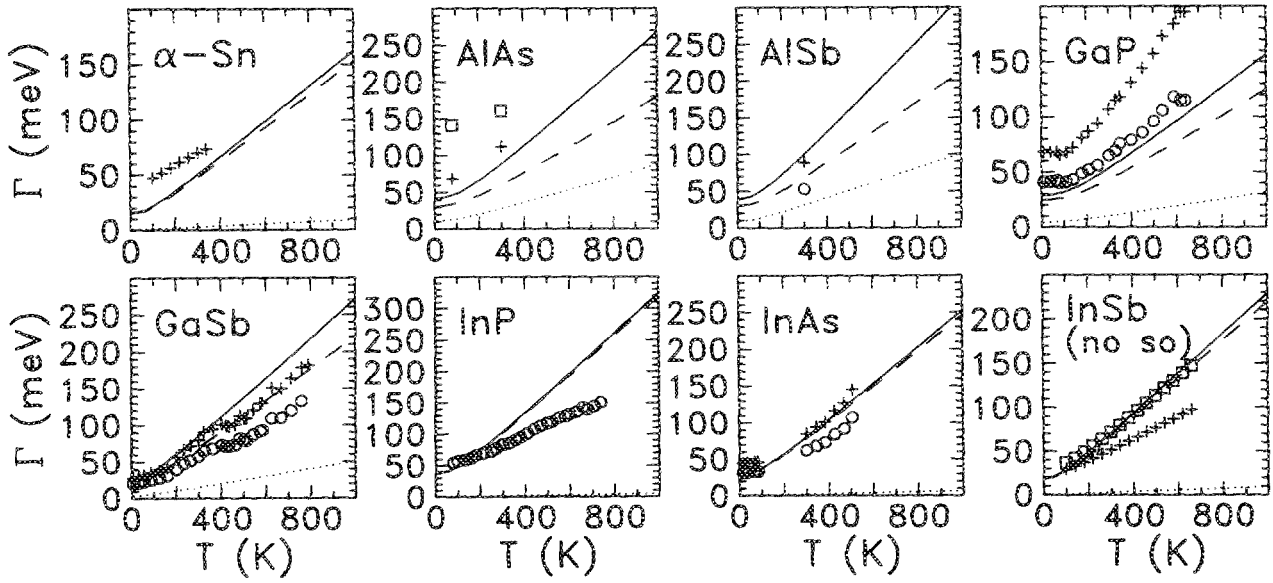

FIG. 2. Calculated broadenings of the E_1 gaps in α-tin, AlAs, AlSb, GaP, GaSb, InP, InAs, and InSb (solid lines), without spin-orbit interaction. Separate contributions from the conduction (dotted lines) and valence bands (dashed lines) are also shown. The experimental broadenings (symbols) are obtained from a line shape analysis with and/or without inclusion of excitonic effects. E_1(2-D)(○), E_1(excitonic)(+), $E_1 + \Delta_1$(excitonic)(□).

TABLE I. Energy loss rates (ELR) for electrons (in units of meV/ps) and holes (meV/fs) at four points in the Λ direction for several diamond and zinc blende semiconductors, calculated from Eq. (3). The averages over Λ and the sum of the ELRs for electrons and holes (in meV/fs) are also shown. Experimental values (in meV/fs), obtained from the broadenings Γ_0 of the E_1 critical point at 0 K (in meV) and assuming an effective phonon energy Ω (in meV), are also given. Note that the largest discrepancy between theory (the column labelled "sum") and the experimental ELR is for α-Sn and may be related to the poor quality of the epitaxial layers used in the measurements.

	Holes					Electrons					Sum	Experiment		
	$L/4$	$L/2$	$3L/4$	L	ave	$L/4$	$L/2$	$3L/4$	L	ave		Γ_0	Ω	ELR
Si	1.95	4.08	5.45	6.51	4.50	3561	2389	1245	980	2044	6.54	65	62	12.25
Ge	0.85	2.03	2.73	3.40	2.25	800	390	27.4	0.00	305	2.56	40	36	4.38
α-Sn	0.40	0.79	1.25	1.54	1.00	70.0	54.6	8.22	2.93	33.9	1.03	45	24	3.28
AlAs	1.38	3.15	4.29	5.48	3.58	1889	1179	568	400	1009	4.59	65	45	8.89
AlSb	0.96	2.37	3.35	5.10	2.95	1234	747	323	192	624	3.57	···	35	···
GaP	1.61	2.65	3.29	4.92	3.12	851	424	205	153	408	3.53	45	45	5.47
GaAs	1.36	2.93	3.98	6.12	3.60	469	377	34.5	1.26	220.4	3.82	30	32	2.92
GaSb	0.66	1.69	2.30	2.93	1.90	57.1	120	29.7	10.8	54.4	1.95	25	28	2.13
InP	1.59	3.37	4.09	4.97	3.51	7.84	65.3	17.1	4.11	23.6	3.53	40	39	4.74
InAs	0.85	1.99	2.37	3.79	2.25	18.5	72.9	26.9	8.15	31.6	2.28	38	27	3.12
InSb	0.42	0.88	1.38	1.68	1.09	12.7	73.2	18.6	10.3	28.7	1.12	30	22	2.01

orbit interaction has been taken into account, for the first time to our knowledge.

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