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TEMPERATURE DEPENDENCE OF THE OPTICAL CONSTANTS OF Ge, α-Sn, AND InSb: ELECTRONIC INTERBAND TRANSITIONS

L. Viña, S. Logothetidis[+], and H. Höchst[++]
Max-Planck-Institut für Festkörperforschung
Heisenbergstrasse 1, D-7000 Stuttgart 80
Federal Republic of Germany

We present dielectric function spectra for Ge, InSb, and α-Sn. These data yield accurate critical point parameters of interband transitions as a function of temperature.

The temperature dependence of the optical constants of all diamond and zinc-blende semiconductors is similar [1]: Features in these spectra shift and broaden with increasing T. These effects have been mainly studied in the region of transparency and near the lowest absorption edge [2]. We report here measurements of the optical constants of Ge, α-Sn, and InSb in the energy range from 1.25 eV to 5.6 eV from 100 K to 850 K (Ge), to 700 K (InSb), and to 350 K (α-Sn). They were performed with an automatic rotating analyser ellipsometer [3]. The critical point (cp) parameters (strengths, Lorentzian broadenings Γ, critical energies, and phase shifts) are obtained through analysis of numerical second derivative spectra of the complex dielectric constant with respect to the photon energy. A shift of the cp to lower energies and an increase in their Γ's, as well as a decrease of excitonic interaction [4] with increasing temperature have been observed.

The real (ε_1) and imaginary (ε_2) parts of the dielectric constant of Ge, α-Sn, and InSb are shown for two different temperatures in Figs. 1 to 3. In the case of Ge and InSb the samples were etch-polished with Syton and a bromine-methanol solution [5]. They were mounted in a windowless cell in flowing dry N_2 and were etched so as to optimize ε_2 at the E_2 critical point [5]. Spectra were measured immediately after etching. The samples were transferred into a ultra-high vacuum dewar. During the measurements the vacuum was 3×10^{-8} torr or better. The α-Sn sample was a 950 Å film grown on a 7mm x 7mm (001)-InSb surface by MBE [6]. The cleanness of the film was controlled by photoemission spectroscopy, there was no indication of surface oxidation. A simple two-phase model [7] was used to evaluate ε from the complex reflectance data in the case of α-Sn. After the transfer of the Ge and InSb samples to the cryostat, oxide layers are still present. We estimate the thickness of these films to be ~10 Å by fitting room temperature data obtained from the sample in the cryostat to the data measured immediately after etching in the N_2-flushed chamber. We assumed that the undesired layers were native oxides with dielectric constants taken from [8,9]. A three-phase model [7] was used for the treatment of data.

[+]On leave from: Aristotle University of Thessaloniki, First Laboratory of Physics, Thessaloniki, GREECE

[++]Present address: Phys. Dept., University of Wisconsin, Madison, Wisconsin 53706,USA

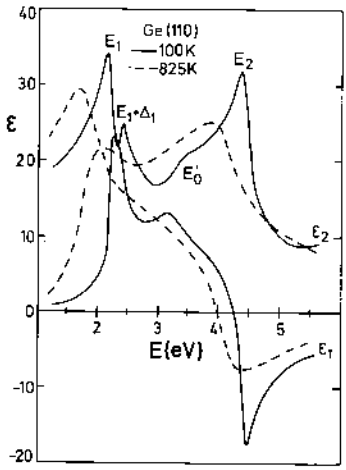

Figure 1 : Dielectric constant for Ge at 100 K and 825 K.

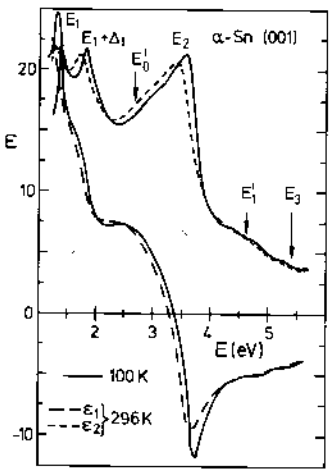

Figure 2 : Dielectric constant for α-Sn at 100 K and 300 K.

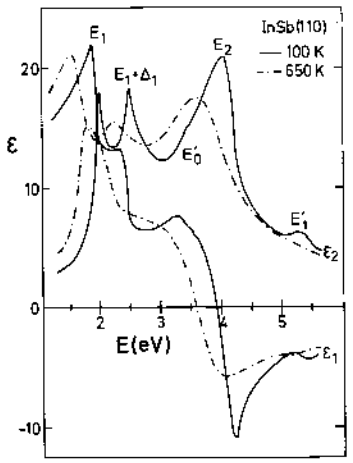

Figure 3 : Dielectric constant for InSb at 100 K and 650 K.

To obtain cp parameters and resolve structure we compute the $d^2\varepsilon/d\omega^2$ spectra from our ellipsometric data. The line shapes of the structures have been fitted to standard expressions for various types of cp. Excitonic effects were taken into account allowing for a mixture of two cp [4]. Two- or three-dimensional cp can be represented by [10]:

(two-dimensional) $\varepsilon \sim C - \ln(E-\omega-i\Gamma)e^{i\phi}$ ($\phi=0$ for minimum; $\phi=\frac{\pi}{2}$ for saddle point) (1)

(three-dimensional) $\varepsilon \sim C - (\omega-E+i\Gamma)^{1/2}e^{i\phi}$ ($\phi=0$ for M_1; $\phi=\frac{\pi}{2}$ for M_2). (2)

The experimental $d^2\varepsilon_1/d\omega^2$ (black dots) for α-Sn at 100 K is displayed in Fig. 4 together with the best fits to cp's for the real and imaginary parts. Similar spectra have been obtained for Ge and InSb. Fine structure that could not be seen in the original spectra are clearly recognized in Fig. 4. Two-dimensional line-shapes have been found to give the best representation for the E_1, E_1', and E_2 cps, three-dimensional

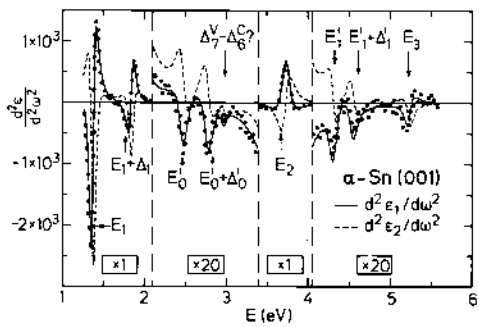

Figure 4 : Second derivative spectra of ε for α-Sn at 100 K. Black points experimental $d^2\varepsilon_1/d\omega^2$. Lines best fits to $d^2\varepsilon_1/d\omega^2$ and $d^2\varepsilon_2/d\omega^2$.

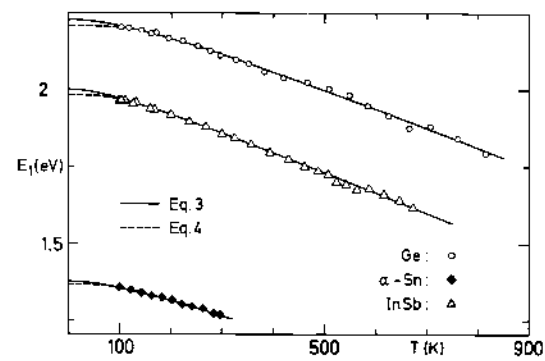

Figure 5 : Energies of the E_1 critical points of Ge, α-Sn, and InSb vs. T. Lines best fits with Eqs. (3) and (4).

line-shapes have been used for the E_0' cps. Figure 5 shows the energy position obtained from our fit to the E_1 transitions in Ge, α-Sn, and InSb as a function of temperature. All results show a linear dependence on temperature at high temperature and a quadratic one at low temperature. These dependences were analysed with two models: Varshni's relation [11] and an expression based on the Bose-Einstein statistical factor which takes into account electron-phonon interaction with phonons of an average frequency Θ:

$$E(T) = E(0) - \frac{\alpha T^2}{\beta+T} \text{ (Varshni)} \tag{3}$$

$$E(T) = E_B - \alpha_B \left[1 + \frac{2}{e^{\Theta/T}-1}\right] \text{ (Bose-Einstein).} \tag{4}$$

Both fits are equally good, differences begin at temperatures lower than our experimental ones. The Debye temperature as well as the mean frequency of phonons taking part in the scattering process decrease in the series Ge; α-Sn; InSb [2]. If the parameters β and Θ from our fits are related to these quantities, they should decrease by the same amount. This is borne out by the experiments: a systematic decrease of these parameters is found: Θ(Ge) = 360(120)K; Θ(α-Sn) = 340(90)K; Θ(InSb) = 272(90)K for the E_1 cp. A similar trend is found for the β parameter and for the other cp's.

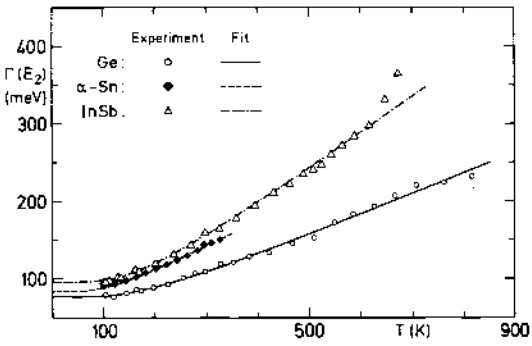

Figure 6 : Dependence of the E_2 cp broadening parameter on T^2 for Ge, α-Sn, and InSb. Lines best fit with Eq. (5).

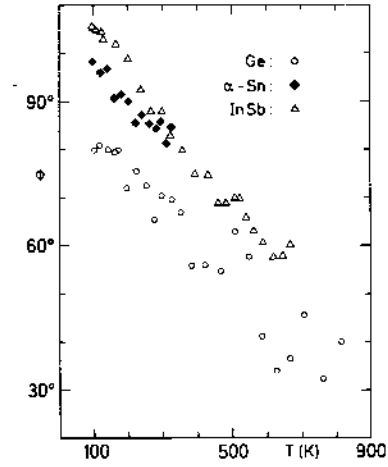

Figure 7 : Dependence on T of the excitonic parameter φ defined in Eqs. (1) and (2) for the E_1 cps of Ge, α-Sn, and InSb.

We show in Fig. 6 the temperature dependence of Γ for the E_2 transitions in Ge, α-Sn, and InSb. The solid lines give best fit to the data with:

$$\Gamma = \Gamma_0\left(1 + \frac{2}{e^{\Theta/T}-1}\right) + \Gamma_1, \tag{5}$$

A similar behaviour of Γ with increasing temperatures is found for the three materials.

The phase angles φ, representing the mixture of contiguous cps are plotted in Fig. 7 versus T for the E_1 transitions in Ge, α-Sn, and InSb. φ decreases rapidly with increasing T: thus excitonic effects decrease. This angle increases through the sequence Ge, α-Sn, InSb, indicating a corresponding increase of electron-hole Coulomb interaction.

Two mechanisms are responsible for the temperature dependence of energy bands at constant pressure [1]: thermal expansion and renormalization of band energies by electron-phonon interaction. The first term can be easily calculated from the dependence of the band structure on volume. Theoretical interest has focused recently on the effect of the electron-phonon interaction. This effect can also be divided into two contributions: Debye-Waller terms [12] and "self-energy" terms [13]. Although reasonable agreement with experiment has been obtained in some work considering only the first contribution, it has been shown that both terms should be included [14]. We can compare our results for the E_0' gap in Ge with full band structure calculations of Debye-Waller and self-energy terms in the T-dependence of the electronic states [15]: a linear fit to the data obtains a T coefficient of $-1.8\ 10^{-4}$ eV/K; assuming that the T-dependence of the Γ_{15} state in Ge is the same as in Si, the E_0' gap in Ge should have a theoretical linear temperature coefficient of -2.2×10^{-4} eV/K (see Fig. 6 in Ref. 15) in good agreement with our experiments. The large values of Θ obtained from our fits indicate that acoustic phonons tend to contribute less than the optical ones, a fact which is also borne out by calculations for the fundamental gap of Ge [15]. More details of the analysis of the other structures present in the spectra will be reported elsewhere [16-18].

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