

Proceedings of the XXVI International School of Semiconducting Compounds, Jaszowiec 1997

VIBRATIONAL MODES IN $\text{Si}_x\text{Ge}_{1-x}$ ALLOYS: TEMPERATURE AND COMPOSITIONAL DEPENDENCE

M. LORENC AND J. HUMLÍČEK

Faculty of Science, Department of Solid State Physics, Masaryk University
Kotlářská 2, 611 37 Brno, Czech Republic

We report infrared absorption spectra of crystalline $\text{Si}_x\text{Ge}_{1-x}$ alloys with silicon content $0 \leq x \leq 1$ at room and liquid nitrogen temperature. We covered the spectral range from 375 to 1200 cm^{-1} that includes the "Si-Ge" and "Si-Si" single-phonon transitions, the continuum of two-phonon processes, and the localized mode of interstitial oxygen. We study the change of vibrational structure and correlation between reference (pure Si and Ge) and alloy spectra. We observed shifts to lower wave numbers by about 1 cm^{-1} of the two-phonon absorption bands per 1% increase in germanium concentration. Pronounced changes of the vibrational spectra upon lowering the temperature were detected.

PACS numbers: 78.30.Am, 63.20.Dj

The $\text{Si}_x\text{Ge}_{1-x}$ alloys rank among the most interesting solid solutions. They represent a continuous series of crystalline materials with gradually varying properties. Vibrational states of the $\text{Si}_x\text{Ge}_{1-x}$ alloys have been studied several times by infrared methods [1-3].

In this paper, we present infrared absorption spectra of bulk $\text{Si}_x\text{Ge}_{1-x}$ alloy for $0 \leq x \leq 1$ in the spectral range 375-1200 cm^{-1} . As far as we know, it is for the first time that the local vibrations of the interstitial oxygen in the alloys are reported. The measurements have been performed on crystalline and polycrystalline samples grown by Czochralski method. The series of 19 samples covered the whole compositional range from Si to Ge, being mostly polycrystalline. Transmittance spectra have been measured with a FTIR spectrometer Bruker IFS55, the spectral resolution was 1 cm^{-1} and a number of scans was typically about 100. The transmission T of an optically thick slab of thickness d is [4]

$$T = \frac{16(n^2 + k^2)}{[(n+1)^2 + k^2]^2} \frac{\exp(-Kd)}{1 - R^2 \exp(-2Kd)} \approx \frac{(1-R)^2 \exp(-Kd)}{1 - R^2 \exp(-2Kd)}, \quad (1)$$

where $K = 4\pi\nu k$ is the absorption coefficient and $R = [(n-1)^2 + k^2]/[(n+1)^2 + k^2] \approx [(n-1)^2]/[(n+1)^2]$ is the reflectivity of a semi-infinite sample with complex refractive index $n+ik$; n is the real part of the refractive index and k is the extinction coefficient. The approximations are valid for $n \gg k$. The real part of the refractive index of the $\text{Si}_x\text{Ge}_{1-x}$ alloy has been computed by

using the formula $n = n_0 + n_1\nu^2$, where $n_0(x) = 4.01 - 0.81x + 0.22x^2$ and $n_1(x) = (0.141 - 0.137x + 0.058x^2)10^{-8} \text{ cm}^2$ [5].

Our samples were nominally intrinsic; the free carrier concentration at room temperature was typically below 10^{15} cm^{-3} . Consequently, the absorption spectra do not show any free-carrier background typical of the earlier data [1]. On the other hand, the scattering of the probing beam on compositional inhomogeneities prevents a part of the transmitted light from reaching the detector. Thus, the observed transmittance in the spectral region of negligible absorption above $\approx 1500 \text{ cm}^{-1}$ is usually lower than the prediction of Eq. (1), inserting $k = 0$ and the proper values of the refractive index n which determine the reflection losses of the thick slabs. Since we did not attempt any correction of the scattering in evaluation of the absorption spectra from the transmittances, it contributes a flat spectral background of different magnitude in the data shown below. However, the scattering background does not influence the target values of the band positions and widths.

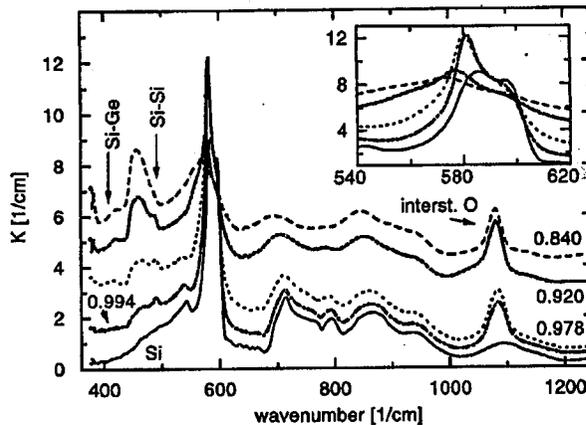


Fig. 1. Infrared absorption spectra of silicon-rich $\text{Si}_x\text{Ge}_{1-x}$ at room temperature. Pure Si sample was grown by the float zone technique and the concentration of interstitial oxygen is negligible. Inset: the dominant TO+TA band on an expanded scale.

We show in Fig. 1 the compositional evolution of the results for silicon-rich alloys. The spectra display several two-phonon combination bands, related to similar structures in pure Si and Ge. In addition, two distinct bands are due to the single-phonon absorption processes, which are allowed due to the loss of symmetry in the alloy; we denote them as “Si-Si” and “Si-Ge” bands, respectively [3]. Our Czochralski samples show also the absorption band of interstitial oxygen at $\approx 1100 \text{ cm}^{-1}$, superposed on a two-phonon background; the overlapping structures were studied earlier in pure Si [4]. Note that the reference Si spectrum of Fig. 1 was obtained with a float-zone silicon, having a negligible oxygen concentration. Upon lowering the temperature, the spectral features shift and sharpen, see Fig. 2.

We have obtained quantitative data on four two-phonon bands and the interstitial oxygen band. We have selected the latter and the TO+TA and TO+LO

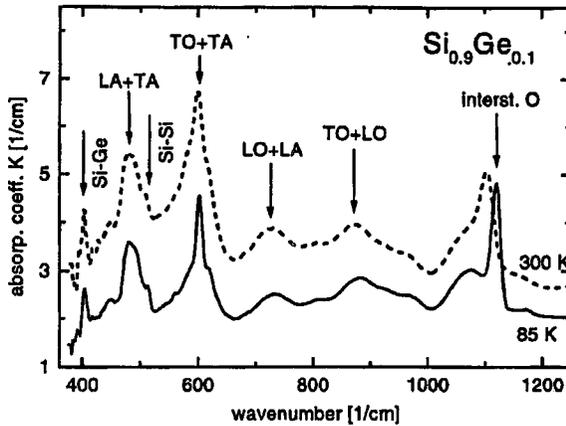


Fig. 2. Infrared absorption spectra of $\text{Si}_{0.9}\text{Ge}_{0.1}$ alloy at room temperature (dashed line) and at 85 K (solid line).

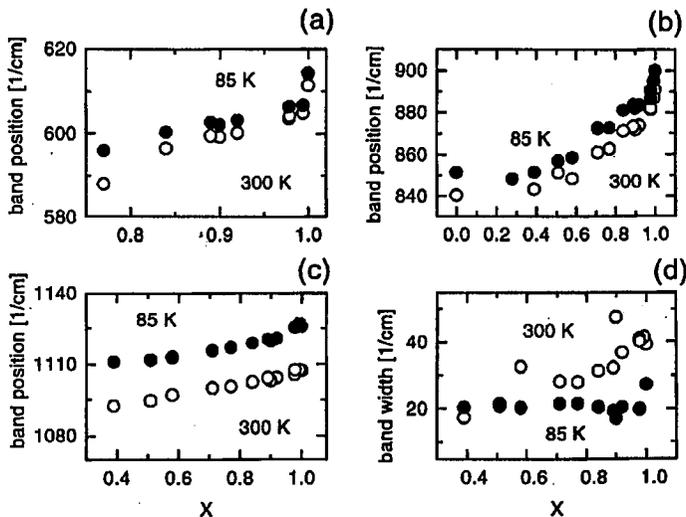


Fig. 3. Compositional dependence of the position of the vibrational bands: TO+TA band (a), TO+LO band (b), interstitial oxygen band (c). The compositional dependence of the band width of the interstitial oxygen band is shown (d). Full circles — 85 K, open circles — room temperature.

combination bands for the detailed presentation here. We list the data in Table and show an overview in Fig. 3. Typical uncertainty is about 0.8 cm^{-1} for the band positions and about 3 cm^{-1} for the widths. The detailed description of the vibrational structures is the main result of the present work. The most pronounced effect of alloying on the band positions consists in the down-shifting with the slope of $\approx 0.7 \text{ cm}^{-1}$ for the TO+TA and TO+LO bands per 1 percent increase in the

TABLE

Temperature and compositional dependence of positions (pos.) and of band width (wd.) of several absorption bands. All data are in cm^{-1} .

	Interst. O				TO+TA				TO+LO	
	300 K		85 K		300 K		85 K		300 K	85 K
x	pos.	wd.	pos.	wd.	pos.	wd.	pos.	wd.	pos.	pos.
1 (Si)	1107.2	39	1125.7	27	611.4	30	614.3	29	890.6	899.7
0.994	1107.5	41	1126.0	21	604.8	31	606.7	24	886.7	894.3
0.979	1107.5	40	1125.4	20	604.1	34	606.4	26	881.8	886.3
0.978	1105.5	41	1125.3	20	603.5	35	606.3	23	881.2	890.4
0.920	1104.3	37	1118.7	20	600.1	50	603.1	34	873.6	883.4
0.900	1103.1	48	1119.6	17	599.2	47	602.1	21	871.7	881.9
0.890	1104.2	32	1120.4	19	599.5	49	602.6	29	873.1	883.4
0.840	1102.5	31	1118.7	20	596.5	55	600.3	40	871.1	880.8
0.770	1100.6	28	1116.8	22	588.0	16	596.0	20	862.6	872.5
0.710	1099.9	28	1115.6	22	569.4	70	577.2	19	860.9	876.5
0.580	1096.9	33	1112.8	20					848.5	858.5
0.510	1094.5	21	1111.7	22					851.2	857.0
0.390	1092.3	17	1110.8	20					843.4	851.4
0.280									848.4	848.2
0 (Ge)									840.4	851.4

Ge content in the middle of the concentration range. Note the steeper decrease in both combination bands for low concentrations of Ge in Si.

The spectral structures observed at low temperatures are shifted to lower energies with respect to the room temperature case. This is consistent with the expected stiffening of the bonds. The Si-O bonding is significantly more influenced, as seen from the larger shift in Fig. 3c. The full account of the results is beyond the scope of the present paper and will be published elsewhere.

In conclusion, we have presented the detailed compositional and temperature dependence of the infrared spectra of $\text{Si}_x\text{Ge}_{1-x}$, including the localized vibrations of the interstitial oxygen impurity.

References

- [1] R. Braunstein, *Phys. Rev.* **130**, 879 (1963).
- [2] A.E. Cosand, W.G. Spitzer, *J. Appl. Phys.* **42**, 5241 (1971).
- [3] S.C. Shen, M. Cardona, *Solid State Commun.* **36**, 327 (1980).
- [4] J. Humlíček, K. Vojtěchovský, *Phys. Status Solidi A* **92**, 249 (1985).
- [5] J. Humlíček, A. Röseler, T. Zettler, M.G. Kekoua, E.V. Khoutsishvili, *Appl. Opt.* **31**, 90 (1992).