

ON FREQUENCY DEPENDENCE OF THE GRÜNEISEN CONSTANT

A.A. Galkin, V.M. Svistunov, O.I. Chernjak and M.A. Belogolovskii

Physico-Technical Institute, Ukrainian Academy of Sciences, 340048, Donetsk, U.S.S.R.

(Received 8 June 1973 by E.A. Kaner)

The different concentration Pb-In alloys were investigated under pressure by the electron tunneling method. The effect of the frequency dependence of the Grüneisen constant determined according to the phonon characteristic frequency shift was displayed.

FOR THE description of the unharmonic effects in solids one should know the dependence of the crystal lattice own frequencies from the volume V . Usually it is considered¹ that the volume change $\Delta V/V$ leads to the relative frequency change $\Delta\omega/\omega$ the same for all modes:

$$\frac{\Delta\omega}{\omega} = \gamma \frac{\Delta V}{V}, \quad (1)$$

where γ is the Grüneisen constant. Such assumption was experimentally confirmed, up to some extent, at least in those experiments where an integral result of the vibrational spectrum was exhibited.

For the first time the tunneling technique enabled us to separate the transverse and longitudinal branches in the vibrational lattice spectrum by the most direct way and the tunneling experiments under pressure gave the possibility to check the justice of the equation (1) for some pure metals. In these experiments the value $\gamma^* = d \ln \omega_{t,i} / dp$ for the characteristic frequencies of the phonon density of states was measured. The calculation results² based on the assumption of γ constancy were in good agreement with the experiments.^{3,4}

The $\text{Pb}_{93}\text{In}_7$ alloy in which the light indium impurity leads to the appearing of the local vibration ω_L in frequency spectrum $F(\omega)$ was studied.⁵ For such an object of the investigation in the range up to 9 kbar we found that $\gamma_{t,i,L}^* = \text{const}$; it was also confirmed by the theoretical conclusions based on

the assumption about small concentration isotopic impurity. However, the real impurity changes not only the atom mass in the corresponding site but also the force constants of the atom interactions that should be especially essential for alloys with the element concentrations close in values.

The present paper reports the results obtained in an investigation of a hydrostatic compressure influence on the phonon spectrum characteristic frequencies of the Pb-In system with In-12% concentration and 40 at.%. We shall not decide upon the experiment details as their descriptions are given earlier.^{3,5} We shall only emphasize that in each measuring cycle two substrates each of which having three samples with different concentration alloys were studied simultaneously. Such experimental technique has excluded the possibility of the errors in pressure measurement when the results for different alloys were compared. The pressure range was 9-11 kbar at which the aluminium remained as superconducting.

The copies of the second-harmonic voltage for different concentration Pb-In samples demonstrating the shift of the corresponding features under pressure are given in the figure. All our numerical results and data of the work⁶ having been received at $p \sim 3.5$ kbar by using solid helium with N-I-S tunneling systems are given in the table. It is evident that the speed $d \ln \omega_{t,i} / dp$ for the main alloy frequencies is slightly changed in comparison with pure lead^{3,4}

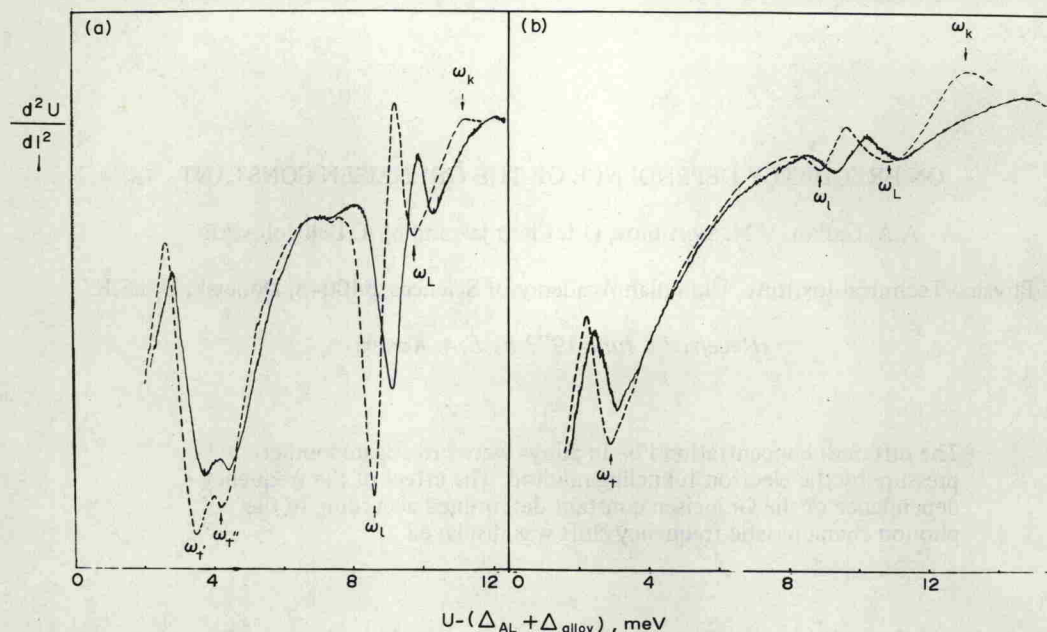


Fig. 1. $(d^2U/dI^2) - U$ characteristics at $P = 0$ (the dashed line) and $P = 9.3$ kbar, $T \sim 1.2^\circ$ K. (a) $\text{Pb}_{88}\text{In}_{12}$; (b) $\text{Pb}_{60}\text{In}_{40}$.

Table 1. Pressure-induced changes in the energy gap and phonon frequencies of Pb-In alloys

X	$2\Delta_0$	ω_t'	ω_t''	ω_L	ω_L	ω_R
$\text{Pb}_{88}\text{In}_{12}$ (meV)	2.68	3.48	4.25	8.61	9.73	11.2
$\text{Pb}_{60}\text{In}_{40}$ (meV)	2.44	2.9		8.73	10.72	13
$\frac{d \ln X}{dP}$ (10^{-6} bar^{-1})	-8.8 ± 0.6	9.1 ± 1	7.2 ± 1	6.2 ± 0.5	5.6 ± 0.5	7.8 ± 1.3
$\text{Pb}_{88}\text{In}_{12}$			7.38 ± 2.94^6	6.13 ± 0.69^6	4.78 ± 1.06^6	
$\frac{d \ln X}{dP}$ (10^{-6} bar^{-1})	-7.9 ± 0.6	7.5 ± 1		5.6 ± 0.7	2.7 ± 0.7	12.5 ± 1.5
$\text{Pb}_{60}\text{In}_{40}$						

while for the impurity zone $d \ln \omega_L / dP$ decreases with an indium concentration increase. The similar tendency which is also appreciable at small concentrations was mentioned in reference 5. The phonon spectrum end is also essentially changed.

One more effect which becomes apparent at small indium concentrations must be emphasized. In this case the impurity zone is asymmetrical; it is connected with the presence not only the single In

atoms surrounded by lead atoms but also with the impurity atom clusters which vibrational frequencies are more than the value ω_L .⁷ Both the width and asymmetry of the curve $d^2U/dI^2 - U$ in the region ω_L are essentially decreased under pressure. It may be as an indication of γ^* -value change even in the limits of the impurity zone. The further addition of indium leads to the expansion of this zone and to the decrease of the above mentioned effect, relatively.