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PHONON INDUCED ANOMALOUS RESISTIVITY IN STRUCTURAL PHASE TRANSITION OF PbSnTe

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Abstract.- We report an anomalous electrical resistivity increment $\Delta \rho$ on PbSnTe near the vicinity of the structural phase transition temperature $T_C$. Applying hydrostatic pressure lowers $T_C$ and leads to a decrease of $\Delta \rho$. It is reasonably interpreted that $\Delta \rho$ is due to the free carrier scattering from the soft TO phonon. The observed initial slope of $T_C$ with increasing pressure is also discussed by using the Littlewood formula.

1. Introduction.- The IV-VI compound semiconductors such as (Pb,Ge,Sn)-Te alloys exhibit a structural phase transition from rocksalt structure to rhombohedrally distorted structure.\(^1\) The softening of the transverse optical (TO) phonon at $q=0$ indicates the structural instability in these small gap materials. A sequence of recent experiments have elucidated the nature of instability associated with the interband electron-phonon coupling.\(^2\) In particular PbSnTe has attracted our attention on the "zero" gap nature as well as on the phase transition. The purpose of present paper is to report the electrical transport measurements on $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ ($x=0.44$, $0.54$, $0.80$ and $0.86$) from $4.2$ to $150$ K by applying hydrostatic pressure. From the anomalous portion of resistivity near $T_C$ under pressure, we explore the pressure dependence of $T_C$ as well as the free carrier scattering mechanism giving rise to $\Delta \rho$.

2. Experiments.- Measurements of Hall coefficient $R_H$ and resistivity $\rho$ were made for solution grown single crystal by using the technique of helium gas compression to produce the hydrostatic pressures. Details of crystal characterization were given elsewhere.\(^3,4\)

3. Results and Analysis.- Since Kobayashi et al.\(^5\) found a resistivity anomaly near $100$ K in SnTe, many authors have investigated the anomalous increase in $\rho(T)$ on PbSnTe\(^3,6\) and PbGeTe\(^6\) near $T_C$. Suski et al.\(^7\) have observed $\Delta \rho$ in PbSnTe by applying pressure. In Fig.1, as a typical example, we show $\rho(T)$ vs. temperature($T$) in Pb$^{0.56}\text{Sn}_{0.44}$Te with carrier concentration $p=2.3 \times 10^{19} \text{cm}^{-3}$ at four different pressures.
Fig. 1: $\rho$ vs. $T$ at four pressures.

There appears a prominent cusp-like anomaly $\Delta \rho$ for each pressure. The lowering peak shift with increasing pressure corresponds to decrease of $T_c$. According to earlier theoretical analysis on SnTe, $\Delta \rho$ in PbSnTe may also be due to the carrier scattering from the soft TO mode. Explicit form of resistivity by the TO phonon scattering $\rho_{TO}$ is given by

$$\rho_{TO} = \frac{2k_F^2}{15pe^2} \frac{2}{k_B} N(E_F) \langle m_{TO}^2 \rangle \int_0^\infty \frac{d\omega}{\omega} \left[ n(\omega_{TO}(k_F\omega)) [1+n(\omega_{TO}(k_F\omega))] \right],$$

where $\omega_{TO}(q)$ is the TO phonon frequency as $\omega_{TO}^2(q) = \omega_{TO}^2(T) + Aq^2$, $k_F$, $N(E_F)$ and $m_{TO}$, respectively, are the Fermi wave number, the density of state at the Fermi level $E_F$ and the matrix element involving the interband electron-phonon deformation potential $\tilde{F}_{CV}$. If we used a high temperature expression for the Planck distribution function as $n(\omega) = k_B T / \hbar \omega$ in Eq. (1), $\rho_{TO}$ is proportional to $T$.

Figure 2 shows the calculated $\rho_{TO}$ by Eq. (1) and the experimental points from Fig. 1 as a function of $T$ at four pressures on Pb$_{0.56}$Sn$_{0.44}$Te. The value of $\tilde{F}_{CV}/\sqrt{A}$ is estimated to be $2.44 \times 10^{-6}$ eV cm$^{-1}$ sec which is obtained by fitting $\rho_{TO}$ with $\Delta \rho$ at 1 bar. The overall feature in $\Delta \rho$ is well reproduced by $\rho_{TO}$ so that $\Delta \rho$ mainly comes from the soft mode scattering. Note that the cusp-like anomaly around $T_c$ in $\rho_{TO}$ is produced by the mean field behavior of $\omega_{TO}$ as
$\omega_{TO}^{2} = \alpha (T - T_{c})$ for $T > T_{c}$ and $\omega_{TO}^{2} = 2 \alpha (T_{c} - T)$ for $T < T_{c}$. In Fig. 3, we plotted $\Delta \rho$ at $T_{c}$; $\Delta \rho(T_{c})$ vs. pressure for all samples. The solid curves denote the calculations by using $\rho_{TO}(T_{c}) = g T_{c}$ with the observed values of $T_{c}$. $g$ is adjusted for each curve to agree with $\Delta \rho$ at 1 bar. We ignored the pressure dependence of the energy gap $E_{g}$, effective mass $m^{*}$ and $A$ which give $g$ in some combination. This approximation may be permitted as long as we are concerned with the carrier concentrations higher than $10^{19}$ cm$^{-3}$ as well as low pressure less than 1 Kbar.

We can see a good correlation between data and calculations. There appears a small discrepancy above 0.5 Kbar which may be attributed to our ignoring of the pressure dependence of $E_{g}$, $m^{*}$ and $A$.

In Fig. 4, we summarized $T_{c}$ vs. tin compositions $x$ at 0.001, 1 and 1.5 Kbar in the four compositions with adding $T_{c} = 98.5$ K in SnTe at 1 bar. When we do not have data just at 1 and 1.5 Kbar, the values estimated by inter- and extrapolation are plotted. The observed initial slope of $T_{c}$ with increasing pressure are estimated to be -14.5, -15.1, -13.4 and -15.0 K/Kbar for $x = 0.44$, 0.54, 0.80 and 0.86, respectively. According to the Littlewood formula, $dT_{c}/dp$ for $x = 0.44$, 0.54, 0.80 and 0.86, respectively, are -12.5, -11.6, -11.8 and -11.9 K/Kbar. The trend and order of magnitude agree well with experiments.

In conclusion we reemphasize that the observed behavior of $\Delta \rho$ under pressure in PbSnTe near $T_{c}$ is well understood by assuming the soft TO phonon scattering with $\omega_{TO}$ described by the mean field scheme.

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