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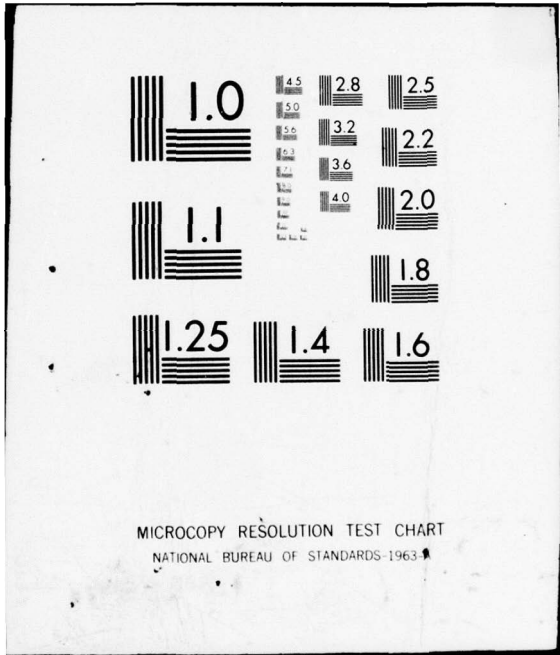
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ELECTRICAL PROPERTIES OF $Pb_{0.8}Sn_{0.2}Te$ EPITAXIAL FILMS

BY J. R. LOWNEY
R. F. BIS
S. C. FOTI

RESEARCH AND TECHNOLOGY DEPARTMENT

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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM	
1. REPORT NUMBER 14 NSWC/WOL/TR-78-5V	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER	
4. TITLE (and Subtitle) 6 Electrical Properties of $Pb_{0.8}Sn_{0.2}$ Te Epitaxial Films		5. TYPE OF REPORT & PERIOD COVERED 9 Final report	
7. AUTHOR(s) 14 J. R. Lowney, R. F. Bis, S. C. Foti		6. PERFORMING ORG. REPORT NUMBER	
9. PERFORMING ORGANIZATION NAME AND ADDRESS Naval Surface Weapons Center White Oak Laboratory White Oak, Silver Spring, Maryland 20910		8. CONTRACT OR GRANT NUMBER(s)	
11. CONTROLLING OFFICE NAME AND ADDRESS		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 27 SR54591501; 0;	
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office) 16 PE 62761N F54591		12. REPORT DATE 11 1 December 77	
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution unlimited		13. NUMBER OF PAGES 17 22 1900	
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		15. SECURITY CLASS. (of this report) UNCLASSIFIED	
18. SUPPLEMENTARY NOTES		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE	
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) $Pb_{1-x}Sn_xTe$, electrical properties, epitaxial Films, thermal strain			
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The temperature dependences of the Hall coefficient and resistivity of $Pb_{0.8}Sn_{0.2}Te$ epitaxial films have been measured from 4.2 to 300K. The results show that no appreciable strain is induced in the epitaxial films grown on BaF_2 substrates because of differential thermal contraction below room temperature.			

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SUMMARY (U)

The research reported herein was carried out in the Solid State Branch of the Materials Division and supported by the Naval Surface Weapons Center, White Oak, Independent Research Fund (IR-725).

J.R. Dixon
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ELECTRICAL PROPERTIES OF $\text{Pb}_{0.8}\text{Sn}_{0.2}\text{Te}$ EPITAXIAL FILMS (U)I. INTRODUCTION

During the past decade the variable band-gap semiconductor $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ has received a great deal of attention because of its use as infrared sensor and laser material. The performance of these devices depends upon the electrical properties of the alloy. The purpose of our work was to investigate the temperature dependence of the Hall coefficient R_H and the electrical resistivity ρ of thin films grown epitaxially on both BaF_2 and KCl substrates. Thin films were selected for this study since their ease of fabrication should lead to their widespread use in future military systems. We have concentrated our study on the alloy $\text{Pb}_{0.8}\text{Sn}_{0.2}\text{Te}$ because this composition is the most widely used.

The transport properties¹⁻⁴ and the band structure⁵⁻⁸ have been studied extensively for x in the range 0.0-0.5. However, many questions still remain unanswered about the scattering of carriers in this range. Ravich *et al*⁹ have shown that for PbTe with carrier densities less than $5 \times 10^{19} \text{cm}^{-3}$ and for temperatures above 10K the predominant carrier scattering mechanisms are acoustic and optic phonons. They also found that in the vicinity of 4.2K impurity and defect scattering predominate.

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In the alloys, however, the carrier mobilities are always lower than those in PbTe¹⁻⁴. For example, at 4.2K the mobility of Pb_{0.8}Sn_{0.2}Te is nearly an order of magnitude lower than comparable PbTe. The carrier mobilities of the films used in our study were comparable to samples^{4,6,10} grown by other methods. Wagner *et al*⁴ have estimated that conventional alloy scattering is not sufficient to account for the large difference in carrier mobility between Pb_{1-x}Sn_xTe and PbTe. One explanation of this lower carrier mobility might be that the crystal structure in the alloys is less perfect than in the pure binary.

We have also investigated the effect of a trigonal strain which could be caused by the difference between the thermal expansion coefficients of the alloy film and its BaF₂ substrate below 300K. BaF₂ has a (111) cleavage plane, and the resulting strains would destroy the cubic symmetry of the film lattice. Evidence exists for this strain in PbTe.¹¹ Since these strains destroy the valley degeneracy, there is a substantial change in the anisotropy factor, defined by neR_H , where n is the carrier density and e is the electronic charge. The strain induced in a film by its KCl substrate does not remove the valley degeneracy because KCl has a (100) cleavage surface, which is an axis of crystal symmetry. Our data indicate that films on BaF₂ with $x = 0.22$ were unstrained.

Another result of our study is that we are able to account for the substantial increase observed in R_H with increasing temperature prior to the onset of minority carrier generation. The explanation of this increase is based primarily on the energy dependence of the electron mass and scattering. Generally it is expected that R_H should be nearly constant so long as a semiconductor is degenerate. However, Pb_{1-x}Sn_xTe is strongly nonparabolic. This leads to an untypically large variation of carrier mobility within a band. As the Fermi distribution smears with increasing temperature, the averages entering into the computation of R_H vary, and the result is a rise in the value of R_H . It is important to understand this phenomenon since variations in R_H can be mistakenly interpreted as changes in carrier density.

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11. Burke, J. R. and Carver, G. P. (to be published in *Phys. Rev.*).

II. EXPERIMENT

A series of thin films of $\text{Pb}_{0.78}\text{Sn}_{0.22}\text{Te}$ were grown on cleaved BaF_2 and KCl substrates by a vacuum evaporation technique.¹⁰ The Sn fractions, x , were determined from measurements of their lattice parameters.¹² The thicknesses were obtained from infrared reflection interference fringes and measured values of the optical dielectric constant.¹³ The carrier densities were determined from a measurement of R_H at high magnetic field, H , ($\mu H \gg 1$), where μ is the carrier mobility. A superconducting magnet swept to 80kOe was used to make these measurements.

The weak field Hall effect ($\mu H \ll 1$) and the resistivity of the samples were measured in a Supravitemp^R research dewar between 4.2 and 300K. The sample configuration was standard¹ and contacts were made with gold pads and silver paint. The temperature was measured with a Au-.07 at % Fe vs chromel thermocouple and stabilized to within 0.1K during the measurements.

The magnetic field was aligned normal to the sample surface, and the resistivity was measured in the plane of the sample. The choice of configuration is important since a strain induced by a BaF_2 substrate would remove the cubic symmetry and valley degeneracy.

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III. THEORY

A. TRIGONAL DISTORTION DUE TO BaF₂ SUBSTRATE

There is a negligible difference between the thermal expansion coefficients of Pb_{1-x}Sn_xTe and BaF₂ above 300K. However, below 300K there may be a strain induced in the film by its substrate because their thermal expansion coefficients become significantly different. To calculate the effect due to strain, we have assumed that the films are free of strain at room temperature and that strain relief does not exist below 300K. The latter assumption leads to an upper limit of the possible induced strain.

A uniform strain, ϵ_{xx} , of 2.0×10^{-3} in the (111) plane at 4.2K was estimated from the thermal expansion coefficients of PbTe¹⁴ and BaF₂¹⁵. The approximate temperature dependence of this strain is given by Equation 1:

$$\epsilon_{xx} = \begin{cases} 2.0 \times 10^{-3}, & T < 25K \\ 2.0 \times 10^{-3} [(T-300)/275]^2, & 25K < T < 300K \end{cases} \quad (1)$$

In order to find the strain normal to the (111) plane, we transformed the stiffness coefficients C_{ij} of PbTe¹⁴ into a coordinate system with the z-axis along the (111) direction. The result is $\epsilon_{zz} = -1.08 \epsilon_{xx}$, which is a contraction nearly equal to the planar expansion. These strains were then transformed into the cubic (100) system to obtain the energy band shifts δE from the theoretical deformation potentials D_{ij} of Ferreira¹⁶:

$$\delta E = \epsilon_{ij} D_{ij} \quad (2)$$

-
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The results at 4.2K are shown in Figure 1. The primary effect is that the shear strains depress the (111) conduction and valence bands below the other equivalent valleys. This trigonal distortion leads to a crystal structure similar to that of bismuth. There are also changes in the energy gaps between the (111) and the other bands as shown in the figure. Note that the values given in Figure 1 are true only for the case of L_6^- conduction and L_6^+ valence bands. After band-inversion these values will invert with the bands. Furthermore, as the temperature increases, the strain relaxes according to Equation (1). Therefore, considerable carrier redistribution will result with increasing temperature. Such a redistribution has important consequences in the temperature dependence of the transport properties of the material.

B. TRANSPORT QUANTITIES

R_H and ρ may be evaluated according to Boltzmann transport theory. However, in these alloys the scattering mechanisms are still not fully understood, and a direct integration of the Boltzmann equation with variational techniques¹⁷ is not possible. In order to obtain tractable expressions for numerical evaluation, we have used the relaxation time approximation (RTA). In addition to employing the RTA we considered the scattering time τ to be an adjustable parameter. At temperatures well below the Debye temperature (130K), inelastic scattering may be expected, in which case the RTA would be invalid. However, we feel that τ , if considered as an effective scattering lifetime, should still adequately represent the scattering.

We have restricted our calculations to the conductivity elements $\sigma_{xx} = \sigma_{yy}$ and σ_{xyz} since the resistivity is measured in the plane of the film and the magnetic field is perpendicular to this plane. In terms of these elements,

$$\rho = 1/\sigma_{xx} \quad (3)$$

and

$$R_H = \sigma_{xyz}/\sigma_{xx}^2 \quad (4)$$

17. Howarth, D. J. and Sondheimer, E. H., Proc. Royal Soc. A219, 53 (1953).

(111) VALLEY

($\bar{1}\bar{1}\bar{1}$), ($1\bar{1}\bar{1}$), ($1\bar{1}\bar{1}$) VALLEYS

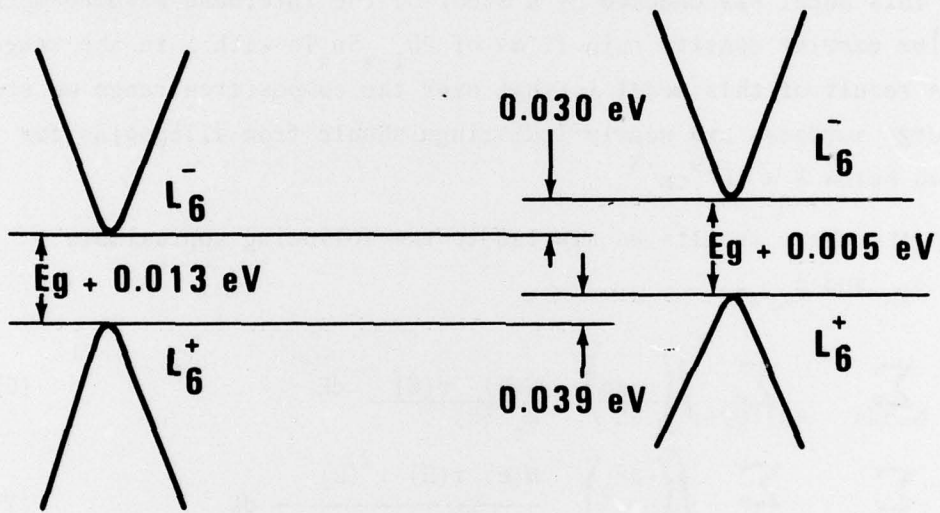


FIGURE 1 SHIFT OF ENERGY BANDS DUE TO STRAIN AT 4.2K

The evaluation of these quantities requires knowledge of the dispersion relation $E(k)$ for both the conduction and valence bands. The energy gap as a function of x is given by Hewes⁸:

$$E_g(x,T) = \begin{cases} 0.187 - 0.543x + 0.02 x^2, & T < 25K \\ E_g(x,25K) + 4.9 \times 10^{-4} (T-25), & T > 25K \end{cases} \quad (5)$$

The dispersion relation is given by the most recent band model for $Pb_{1-x}Sn_xTe$.¹⁸ The validity of this model was checked by a study of the interband magneto-optic reflectance of low carrier density thin films of $Pb_{1-x}Sn_xTe$ with x in the range 0.0 to 0.4.¹⁹ A result of this model is that over the composition range we studied the constant energy surfaces are nearly indistinguishable from ellipsoids for carrier densities below $5 \times 10^{19} \text{ cm}^{-3}$.

Based upon the above results we are led to the following approximate expressions for σ_{xx} and σ_{xyz} :

$$\sigma_{xx} = e^2 \sum_{\text{bands}} \sum_{\text{valleys}} \int \left(\frac{-\partial f_0}{\partial E} \right) \frac{N(E) \tau(E)}{m_c(E)} dE \quad (6)$$

$$\sigma_{xyz} = e^3 \sum_{\text{bands}} \sum_{\text{valleys}} \int \left(\frac{-\partial f_0}{\partial E} \right) \frac{N(E) r(E) \tau^2(E)}{m_c^2(E)} dE \quad (7)$$

where $N(E)$ is the total number of electron states within a constant energy surface E , $m_c(E)$ is the conductivity effective mass, $r(E)$ is the anisotropy factor and $f_0(E)$ is the Fermi-Dirac distribution function. For $\tau(E)$ we have used the following expression:

$$\tau(E) = \left(A(T) \left[d(E) + \frac{C}{v(E)} \right] \right)^{-1}, \quad (8)$$

-
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where $d(E)$ is the density of states and $v(E)$ is the average carrier velocity. The two factors, $d(E)$ and $v(E)$, are representative of the contributions of acoustic or optic phonon scattering, respectively. Although it is not known whether acoustic or optic phonon scattering is dominant in the alloys, our rationale for using Equation 8 is that it represents the admixture of two scattering mechanisms with very different energy dependencies. The first term weights the contribution of carriers near the band edge more heavily by giving them relatively longer scattering lifetimes. The second term weights the contribution of carriers deeper into the band more heavily by giving them proportionately longer lifetimes. Therefore, by varying the constant C , we can investigate the dependence of R_H over a wide range of energy dependences of $\tau(E)$.

A priori the constant C may be considered to be a function of temperature. However, we found that we were able to obtain good fits to our data by making C independent of temperature. Therefore, the entire explicit temperature dependence of the scattering lifetime is contained in $A(T)$. There is also an implicit temperature dependence of $\tau(E)$ because of the variations in $d(E)$ and $v(E)$ with temperature as the energy gap changes.

The temperature dependence of R_H may be examined more readily by considering the following reduced expression:

$$R_H = \frac{p \langle \mu_p^2 r_p \rangle - n \langle \mu_n^2 r_n \rangle}{e (p \langle \mu_p \rangle + n \langle \mu_n \rangle)^2} \quad (9)$$

where n and p are respectively the electron and hole densities, μ and r refer to the mobility and the anisotropy factor, respectively. The symbol $\langle \rangle$ refers to the appropriate average over the band as implied by Equations 6 and 7. At low temperatures, there is only one carrier present and there will be a variation in R_H with temperature resulting from the energy dependence of the mobility and from the possible redistribution of carriers among the valleys as strain relaxes. At higher temperatures, there will be generation of minority carriers which will tend to cancel the majority carrier Hall effect. The amount of cancellation depends on the ratio of the majority to minority carrier mobilities. From previous work on $Pb_{1-x}Sn_xTe^3$, it appears that below 300K, electrons have a mobility about twice that of holes. Therefore, we shall consider the ratio of electron to hole mobility as an adjustable parameter with an expected value close to 2.0.

IV. RESULTS AND DISCUSSION

Plots of the temperature dependence of the resistivity and Hall coefficient of four representative $\text{Pb}_{0.78}\text{Sn}_{0.22}\text{Te}$ samples appear in Figures 2-5. The solid curves are a best theoretical fit to R_H . Below 200K the band parameters of Hewes *et al*⁸ were used. Above 200K, the primary valence-conduction band momentum matrix elements were decreased by 12% to agree with the larger effective masses reported by other workers.^{8,13} The ratio of electron to hole scattering lifetime, which is approximately equal to their mobility ratio because of the band symmetry, was found to be 3.0 in these samples. This value is larger than the value of 2.0 reported in bulk crystals. The constant C, used in Equation 8 was found to be the same for all the samples. For the sample with $p = 1.6 \times 10^{17} \text{ cm}^{-3}$ this constant value of C implies that the scattering is 50% acoustic phonon-like and 50% optic phonon-like below 100K. At room temperature the scattering becomes 80% acoustic phonon-like. The samples with $p = 3.4 \times 10^{18} \text{ cm}^{-3}$ and $3.8 \times 10^{18} \text{ cm}^{-3}$ are limited predominately by acoustic phonon-like scattering with the optic phonon-like accounting only for 10% at all temperatures. In contrast to pure bulk PbTe ,⁹ acoustic-phonon like scattering appears to be stronger in the alloys.

It is the admixture of the relatively more mobile carriers near the band edge which leads to the observed upsweep in R_H . This effect could easily be mistaken as impurities or defects and lead to erroneous interpretation of data. Furthermore, the generation of minority carriers causes R_H to decrease at high temperatures as seen in Figures 2 and 3.

An important conclusion may be drawn by comparing the theoretical curves for R_H with and without strain. The curves for the strain free case fit the data much better than those with strain included. Additional evidence that these films on BaF_2 are unstrained is seen by comparing the data of Figure 5 with that of Figure 4. These two films have nearly the same carrier density, but the sample of Figure 5

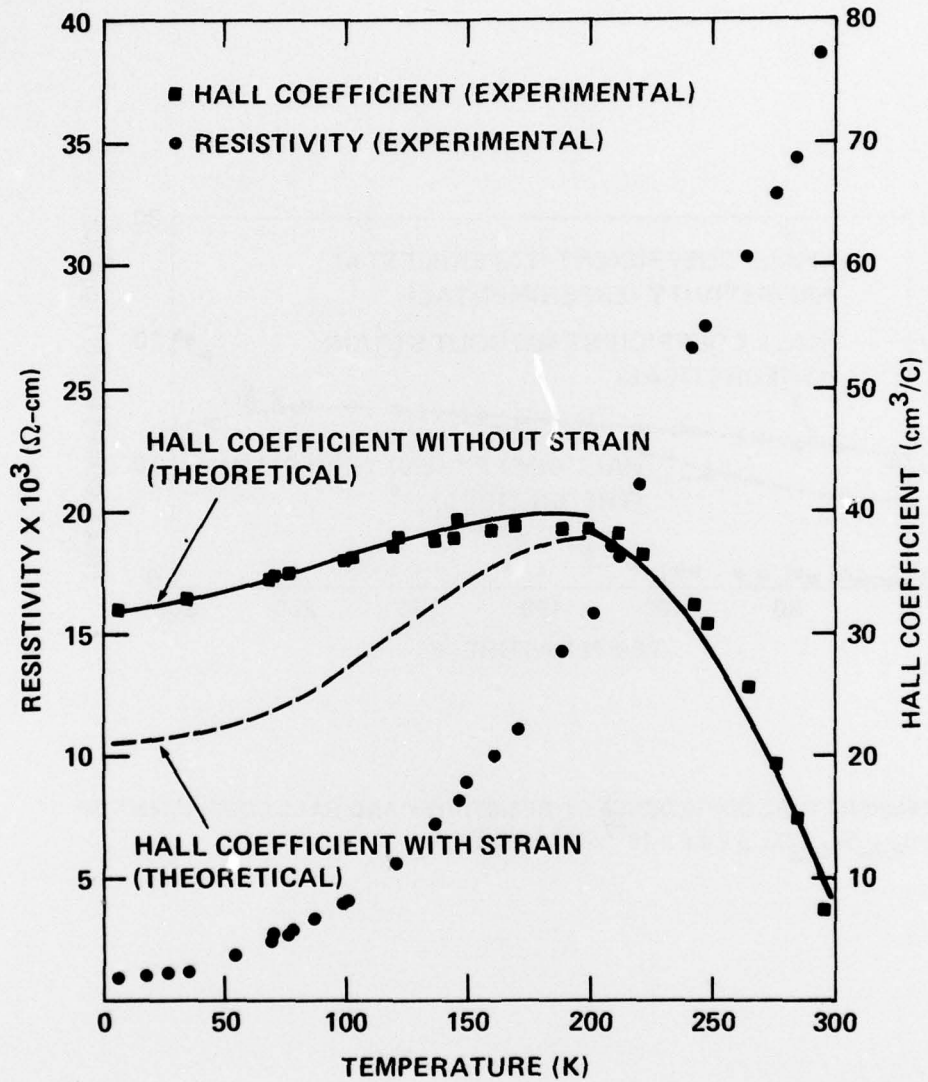


FIGURE 2 TEMPERATURE DEPENDENCE OF RESISTIVITY AND HALL COEFFICIENT OF
 $\text{Pb}_{0.78}\text{Sn}_{0.22}\text{Te}$, $\rho = 1.6 \times 10^{17} \text{cm}^{-3}$, ON BaF_2 .

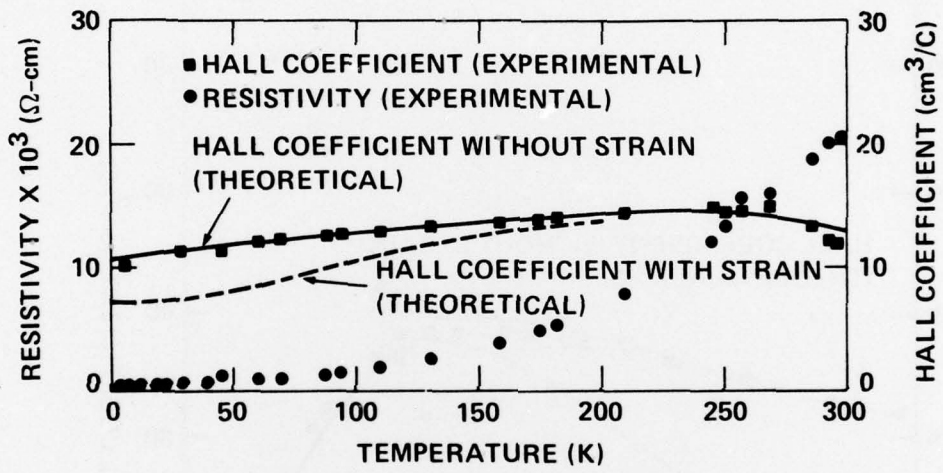


FIGURE 3 TEMPERATURE DEPENDENCE OF RESISTIVITY AND HALL COEFFICIENT OF $\text{Pb}_{0.78}\text{Sn}_{0.22}\text{Te}$, $\rho = 4.6 \times 10^{17} \text{cm}^{-3}$, on ON BaF_2 .

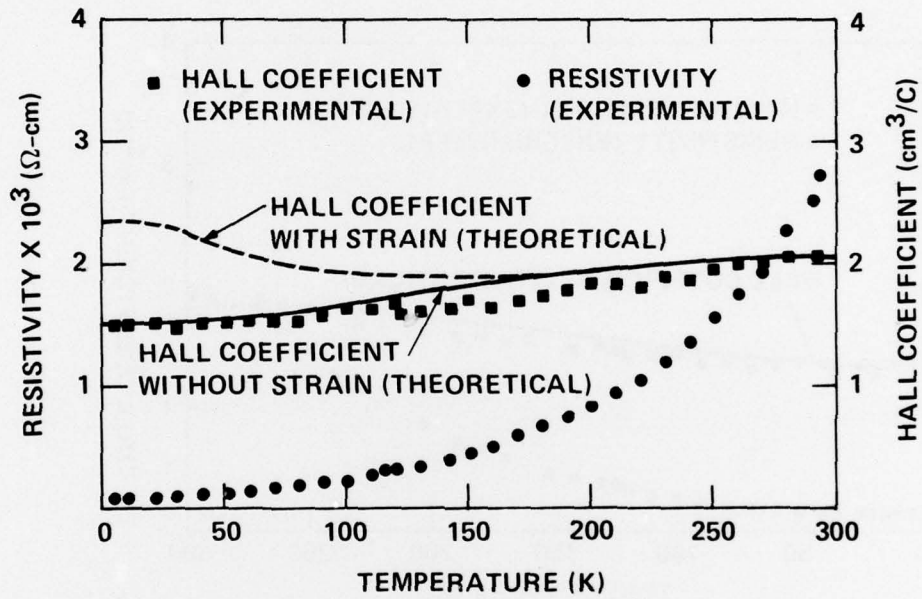


FIGURE 4 TEMPERATURE DEPENDENCE OF RESISTIVITY AND HALL COEFFICIENT OF $\text{Pb}_{0.78}\text{Sn}_{0.22}\text{Te}$, $\rho = 3.4 \times 10^{18} \text{cm}^{-3}$, ON BaF_2 .

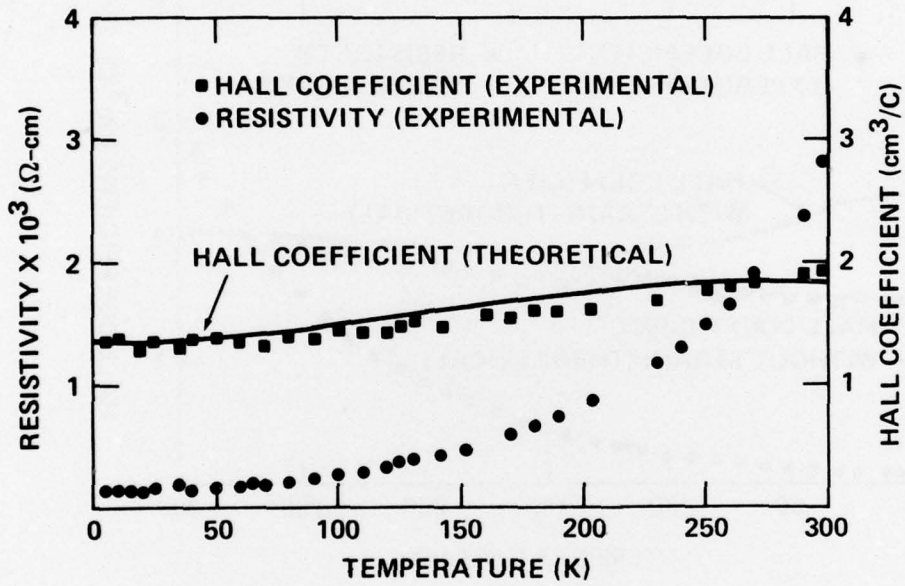


FIGURE 5 TEMPERATURE DEPENDENCE OF RESISTIVITY AND HALL COEFFICIENT OF $\text{Pb}_{0.78}\text{Sn}_{0.22}\text{Te}$, $\rho = 3.8 \times 10^{18} \text{cm}^{-3}$, ON KCl.

was grown on KCl instead of BaF₂. Since any strain induced in a film by KCl does not remove valley degeneracy, there is very little difference between the R_H dependence of such a film and a truly strain free one. The close agreement between the variation of R_H in both figures shows that there must be almost no strain induced in the film on BaF₂.

Let us now consider the resistivity of the four Pb_{0.78}Sn_{0.22}Te samples. We wished to find the explicit temperature dependence of $\tau(E)$, i.e. $(A(T))^{-1}$ of Equation 8. In order to accomplish this we subtracted out the residual resistivity measured at 4.2K and divided out the implicit temperature dependence of $\tau(E)$. The results obtained showed that for the lowest, intermediate and highest carrier density samples A(T) varies approximately as T, T^{3/2} and T², respectively.

V. CONCLUSION

We have measured the temperature dependence from 4.2 to 300K of the Hall coefficient R_H and resistivity ρ of several Pb_{0.78}Sn_{0.22}Te films grown on BaF₂ and KCl substrates. Theoretical fits to the data were obtained by using the most recent band model and treating the scattering lifetime as an adjustable parameter. The possible effects of strain induced in a film by its substrate were also included. Much better fits to our data were obtained if the effects of strain were not included. Comparison between a film on BaF₂ with a film on KCl further confirmed the absence of strain.

ACKNOWLEDGMENT

The authors would like to thank Drs. G. P. Carver and B. B. Houston of this Center for their help in making the measurements at high magnetic field. We are also indebted to Drs. J. R. Dixon and R. S. Allgaier for their helpful discussions.

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