this is done the overlap comes out to be very close to 10^{-2} electron per atom and this is a reasonable figure. Also the fact that the corrected value of $\kappa_\perp$ is small and substantially independent of composition tends to support the correctness of the above views.

It must, however, be pointed out that the behavior of Sb-Pb and Sb-Te alloys are at variance with the theory. The $\kappa_{||}$ for Pb shows no inversion although its valence is the same as that of Sn or Ge. However the value of $\kappa_{||}$ does indeed decrease, as it should, and when it is remembered that the Pb atom (atomic weight 207) is much heavier than the Sb atom (atomic weight 121.8) it is perhaps reasonable to suppose that the structure of the antimony lattice will undergo more modification here than in the case of the lighter elements.

In the case of Sb-Te, $\kappa_{||}$ should increase since in this case we are adding to the overlap because Te possesses one more electron per atom than Sb. This is seen not to be the case, although $\kappa_{||}$ for Sb-Te does decrease much less than for any of the other alloys. Also here the atomic weights in the two cases are very similar so that the argument advanced in the case of Sb-Pb is not tenable.

Despite these imperfections, however, the results on the whole do offer a rather gratifying check on the correctness of the current theory. One of us (S.H.B.) presented part of this work to the Faculty of the Graduate School of Yale University for the degree of Doctor of Philosophy. Part of the equipment used in this work was purchased by a grant from the George Sheffield Fund.

DECEMBER 15, 1941 PHYSICAL REVIEW VOLUME 60

Temperature Variation of the Magnetic Susceptibilities of Antimony-Tin Alloys

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(Received September 8, 1941)

The principal magnetic susceptibilities of single crystals of pure antimony and of antimony-tin alloys up to 4.1 atomic percent tin have been determined in the temperature range 77°K to room temperature. The measurements were made with a modified Sucksmith-Jackson ring balance. It is found that the diamagnetic suspectibility perpendicular to the trigonal axis ($\kappa_\perp$) is practically independent of temperature for all the alloys. The susceptibility parallel to this axis ($\kappa_{||}$) decreases in magnitude with increase in temperature, both for alloys in which $\kappa_{||}$ is diamagnetic and those in which it is paramagnetic, with one exception, the alloy of 1.06 atomic percent tin. A suitable correction for the temperature-independent diamagnetic susceptibility of the lattice ions brings this exception into agreement with the others. The experimental values of $\kappa_{||}$ are compared with Stoner's expressions for the susceptibilities of free electrons, on the assumption that $\kappa_{||}$ is due to the electrons overlapping into the second Brillouin zone. Good agreement is obtained by adjusting the parameter ζ_0, the upper limit of the Fermi distribution, and the degeneracy temperature of these electrons is found to be approximately 475°K, about 0.25 of what it would be for perfectly free electrons. This indicates that the effect of the lattice field is to suppress the value of ζ_0 by increasing the density of electronic energy levels. Values of the effective masses of the electrons, which occur as parameters in the energy equation, are computed, and the energy interval between the bottom of the second Brillouin zone and the top of the Fermi distribution is estimated as about 0.04 ev.

IT has been pointed out in a previous paper[1] that the magnetic susceptibility of a solid should depend chiefly on the concentration of the electrons in the solid and on the temperature. In that paper (referred to hereafter as I) the effect of changing the electron concentration in single crystals of antimony by alloying with small amounts of other metals was discussed. In the present discussion, the effect of temperature variation is to be considered. In particular, the principal magnetic susceptibilities of single crystals of antimony-tin alloys of from zero to approximately 4.1 atomic percent tin

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[1] S. H. Browne and C. T. Lane, Phys. Rev. **60**, 895 (1941).

were determined at temperatures ranging from 77°K to room temperature.

The antimony crystal lattice has two atoms per unit cell, and five valence electrons per atom, so there are just enough valence electrons per unit cell to fill five Brillouin zones. But since only the highest filled or nearly filled zone and the next higher zone are of interest, it is convenient to refer to these as the first and second Brillouin zones, respectively. The results of susceptibility measurements at room temperature, described in I, suggest that in antimony the number of electrons per atom which overlap into the second zone is of the order of 10^{-2}, or about $3.3(10)^{20}$ electrons per cm^3. For a free electron gas, this corresponds to an electron concentration somewhere between those of classical and degenerate gases. Without knowing just how the lattice field will affect these electrons, it is impossible to anticipate their behavior in the crystal lattice.

From the point of view of the modern theory, an electron gas in a solid may exhibit paramagnetism, due to the electron spin, or diamagnetism. For a free electron gas, Stoner[2] has developed expressions for the magnetic susceptibilities which are valid in two limiting cases, the case of a completely degenerate gas, and the case of a non-degenerate gas. Let κ_p denote the paramagnetic and κ_d the diamagnetic susceptibility per unit volume. Then Stoner's formulae may be written as follows:

I. $kT \ll \zeta_0$

$$\kappa_p = \frac{3}{2}\frac{n\mu^2}{\zeta_0}\left\{1-\frac{\pi^2}{12}\left(\frac{kT}{\zeta_0}\right)^2-\frac{1}{6}\left(\frac{\mu H}{\zeta_0}\right)^2\right\}, \quad (1)$$

$$\kappa_d = -\frac{1}{2}\frac{n\mu^2}{\zeta_0}\left\{1-\frac{\pi^2}{12}\left(\frac{kT}{\zeta_0}\right)^2+\frac{1}{10}\left(\frac{\mu H}{\zeta_0}\right)^2\right\}. \quad (2)$$

II. $kT \gg \zeta_0$

$$\kappa_p = \frac{n\mu^2}{kT}\left\{1-\frac{1}{3}\left(\frac{2}{\pi}\right)^{\frac{1}{2}}\left(\frac{\zeta_0}{kT}\right)^{\frac{3}{2}}-\frac{1}{3}\left(\frac{\mu H}{kT}\right)^2\right\}, \quad (3)$$

$$\kappa_d = -\frac{1}{3}\frac{n\mu^2}{kT}\left\{1-\frac{1}{3}\left(\frac{2}{\pi}\right)^{\frac{1}{2}}\left(\frac{\zeta_0}{kT}\right)-\frac{1}{15}\left(\frac{\mu H}{kT}\right)^2\right\}. \quad (4)$$

In these equations, n is the number of electrons per unit volume, μ is the Bohr magneton, ζ_0 is the thermodynamic potential per electron corresponding to the upper limit of the Fermi distribution at the absolute zero of temperature, H is the magnetic field, and k and T are Boltzmann's constant and the temperature. μ and ζ_0 are given by the relations

$$\mu = eh/4\pi mc \quad (5)$$

and

$$\zeta_0 = (h^2/2m)(3n/8\pi)^{\frac{2}{3}}. \quad (6)$$

The terms involving the magnetic field are negligible, since $\mu H \ll kT$ except for very high fields and extremely low temperatures. The terms which give the temperature dependence, and indeed both of the Eqs. (3) and (4), are of

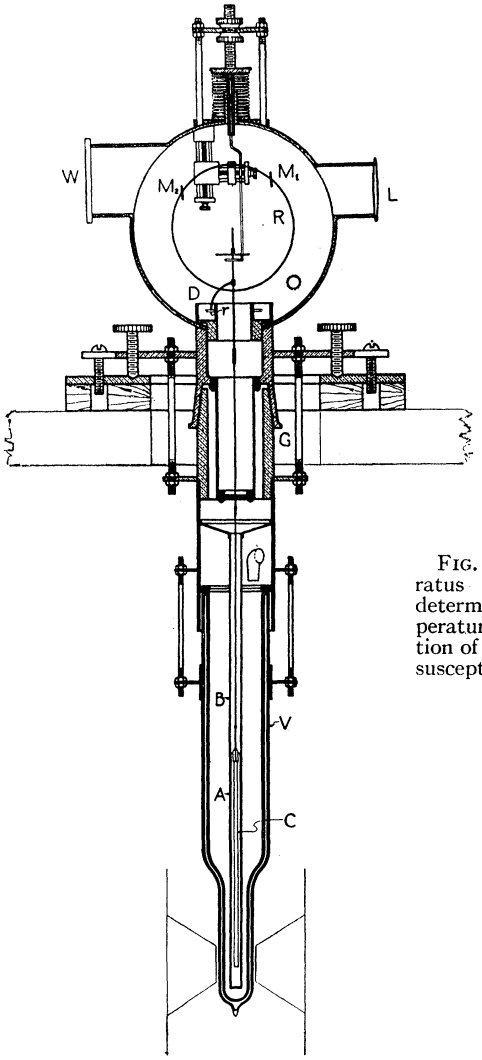

FIG. 1. Apparatus used to determine temperature variation of magnetic susceptibilities.

[2] E. C. Stoner, Proc. Roy. Soc. **A152**, 672 (1935).

little interest unless ζ_0/k is a temperature that can be attained for any particular case. Except for the field dependent parts of Stoner's equations, the resultant susceptibility of the electron gas is the simple sum of the dia- and paramagnetic parts. Thus the resultant susceptibility is paramagnetic, since the magnitude of κ_d is $\frac{1}{3}\kappa_p$.

The electrons in a solid are certainly not free, since their behavior depends on the periodic potential of the lattice field. Before any quantitative expressions can be deduced, it is necessary to know how the electronic energies are affected by the lattice field. The calculation which is involved cannot be carried out in most cases, but it is often possible, in a Brillouin zone which is only partially filled, to represent the energy of an electron by an equation of the form

$$E=(h^2/2m)(\alpha_1 k_x^2+\alpha_2 k_y^2+\alpha_3 k_z^2), \tag{7}$$

where k_x, k_y, and k_z are the components of the wave number vector $\mathbf{k}$, and α_1, α_2, and α_3 are parameters which can be interpreted as the ratios of the electronic mass to the effective mass of an electron, referred to the directions of x, y, and z, respectively. Obviously, Eq. (7) represents an ellipsoidal energy surface in $\mathbf{k}$ space. The origin is at the center of the ellipsoid, and lies in a plane separating the zone under consideration from the next lower one. Near the boundary of a zone, the curvature of an energy surface is great, so the aproximation is likely to give reasonably good results when the number of electrons in the higher zone is small.

The magnetic susceptibilities of the electrons in the lattice are different from those of free electrons. If their energies are given by Eq. (7), the susceptibilities, to a degree of approximation corresponding to the first term in each of Stoner's equations, and for the same limiting cases, are

I. $kT\ll\zeta_0^*$

$$\kappa_p=\frac{3n\mu^2}{2\zeta_0^*}=\frac{3n\mu^2}{2\zeta_0}(\alpha_1\alpha_2\alpha_3)^{-\frac{1}{3}}, \tag{8}$$

$$\kappa_d=-\frac{n\mu^2}{2\zeta_0^*}\alpha_1\alpha_2=-\frac{n\mu^2}{2\zeta_0}\left\{\frac{(\alpha_1\alpha_2)^2}{\alpha_3}\right\}^{\frac{1}{3}}. \tag{9}$$

II. $kT\gg\zeta_0^*$

$$\kappa_p=n\mu^2/kT, \tag{10}$$

$$\kappa_d=-(n\mu^2/3kT)\alpha_1\alpha_2. \tag{11}$$

The quantity ζ_0^* in these equations is the value of ζ_0 for the electrons in the lattice. Its value[3] is

$$\zeta_0^*=\zeta_0(\alpha_1\alpha_2\alpha_3)^{\frac{1}{3}}. \tag{12}$$

The fact that Eq. (8) differs from the first term of (1) is explained by Eq. (12). It is assumed that the spin is unaffected by the lattice forces, and the spin paramagnetism is changed only because of the change in the limit of the Fermi distribution for given n.

It should be noted that the ratio of the magnitude of κ_d to κ_p is now $\frac{1}{3}\alpha_1\alpha_2$. Thus, depending upon $\alpha_1\alpha_2$, the resultant susceptibility may be diamagnetic.

Experimental Procedure

The magnetic measurements at room temperature, described in I, showed that in crystals of alloys of antimony-tin the diamagnetic susceptibility parallel to the trigonal axis, $\kappa_{||}$, decreases rapidly with increasing amounts of tin, and for crystals of greater than 1.17 atomic percent tin the crystals are paramagnetic in this orientation. The greatest paramagnetic susceptibility occurs in crystals of a little more than 4 percent. In order to cover this range, five of the crystals previously used were taken for the temperature dependence measurements. These were of pure antimony and of alloys of 0.49, 1.06, 2.24, and 4.10 atomic percent tin.

It was desired to determine the change of the principal susceptibilities of these crystals with temperature, over a range of temperature from 77°K (liquid nitrogen) to room temperature. For this purpose, one of the most satisfactory methods is that which employs the Sucksmith ring balance,[4] as modified by Jackson.[5] Previous work with this method has been carried out with very small single crystals by Jackson, Schoenberg and Uddin,[6] and others. To use this apparatus with the crystals on hand it was necessary to adapt it to the Gouy method, since these crystals were in the form of rods, about 15 cm long and 0.3 cm diameter.

[3] A discussion of the effect of the lattice field on the electrons in a crystal can be found in N. F. Mott and H. Jones, *Properties of Metals and Alloys*, (Clarendon Press, Oxford, 1932) chapter VI.

[4] W. Sucksmith, Phil. Mag. **8**, 158 (1929).

[5] L. C. Jackson, Proc. Roy. Soc. **A140**, 695 (1933).

[6] D. Schoenberg and M. Zaki Uddin, Proc. Camb. Phil. Soc. **32**, 499 (1936).

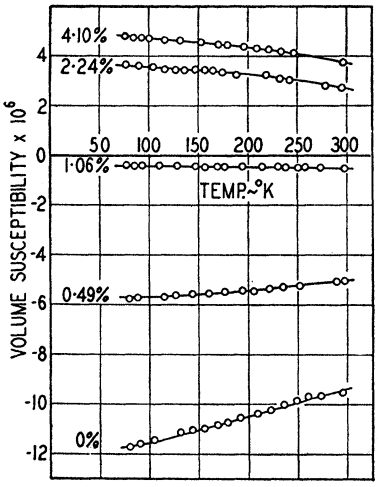

FIG. 2. Temperature variation of $\kappa_{\parallel}$; Sb-Sn alloys.

A sectional view of the apparatus is shown in Fig. 1. Because of the size of the crystals used, it was necessary that the phosphor bronze ring R be made of heavier material than has usually been used with this method, but the larger magnetic force which can be obtained compensates, at least in part, for the loss of sensitivity brought about by this modification. The ring which was used was 9 cm in diameter, and was made of a strip of bronze 0.3 cm wide and 0.015 cm thick.

The crystal C was suspended from the ring by a rigid Pyrex glass fiber, with its lower end between the poles of a Weiss electromagnet. The crystal was surrounded by a copper tube A, connected to the upper housing through a German silver tube B. One junction of a copper-constantan thermocouple was soldered to the copper tube to provide a means of measuring the temperature. The refrigerant, liquid nitrogen, was contained in the Dewar flask V. To damp out vibrations in the suspended system the wire ring r, which moves in the dish of oil, D, was mounted on the suspension.

The motion of the crystal under the action of a magnetic field produces a deflection of a beam of light which passes through the lens L and is reflected off the mirrors M_2 and M_1 out the window W. The source of light was an ordinary galvanometer lamp upon which was mounted a screen containing a narrow horizontal slit. The image of the slit was received by a traveling microscope with which the deflection was measured. As a reference line, one of the narrow bands of the diffraction pattern of the slit was used. To bring out the pattern clearly, an orange-yellow filter was placed before the slit.

If u is the deflection of the crystal and v the corresponding deflection of the image at the microscope, then

$$\frac{v}{u}=\frac{0.943(4D+2d)}{R},$$

where D is the distance from M_1 to the microscope, d the distance between M_1 and M_2, and R the radius of the ring.[7] In this case D was approximately 160 cm, d about 6.5 cm, and R was 4.5 cm. This gives $v/u \backsimeq 135$. Values of v which were obtained with magnetic fields of about 9000 gauss were in most cases of the order of 1 to 3 mm, and were read to the nearest 0.001 mm, though the possible error in a given reading may have been as large as 0.01 mm in the worst cases, because of the difficulty of obtaining a fine image in the microscope. The accuracy of the measurements could be made greater by improving the optical system, but for this particular problem results correct to within about one percent were considered satisfactory.

To eliminate the large magnetic effect of oxygen at low temperatures, the air in the balance housing was pumped out and replaced with hydrogen gas, which is weakly diamagnetic. It was found that with gas pressures much below 25 cm of Hg there was considerable thermal lag between the crystal and the surrounding copper tube, so that the thermocouple would not give the temperature of the crystal if this were

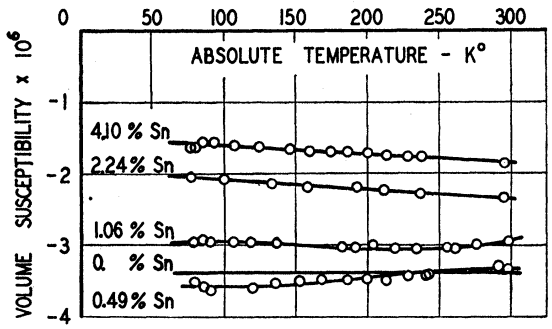

FIG. 3. Temperature variation of $\kappa_{\perp}$, Sb-Sn alloys.

[7] This relation, and those relating the deflections u and v with the force on the crystal and the dimensions of the ring are given by Sucksmith, reference 4.

changing rapidly. Because it was desired to take readings with the temperature changing continuously, a pressure of approximately 65 cm of Hg was maintained in the balance.

The thermocouple was calibrated from a measurement with liquid nitrogen, which was found to agree closely with the value given by Weber[8] for copper-constantan. At all other temperatures Weber's values were used to convert the thermocouple readings to temperatures, computed to the nearest 1°C.

The deflection u is proportional to the magnetic force on the crystal, and hence to the magnetic susceptibility, if the field is kept constant. Since the susceptibilities were known for all the crystals at room temperature, the balance could be calibrated by observing its deflection for a given magnetic field at this temperature. A deflection was observed for each orientation of the crystal, that is, one with the principal axis of the crystal parallel and one with it perpendicular to the magnetic field. Liquid nitrogen was then introduced into the Dewar flask, and when the system had come to equilibrium the new deflections were read, with the same current in the electromagnet as before. The magnet cores were demagnetized between readings to insure the same field for each.

The time required for the crystal to return to room temperature after the liquid nitrogen had evaporated was about 3 hours. During this time observations were made successively with the crystal alternately in one orientation and the other. Since it was not convenient to rotate the crystal, this was accomplished by rotating the magnet back and forth through 90°.

It was noted that there was a gradual change in the position of zero deflection as the temperature changed. This was due partly to the change in density of the hydrogen gas, and partly to other causes. To account for the shift, the position of the zero was checked before and after each reading.

Results

The results of the measurements are shown in Fig. 2 and Fig. 3. In Fig. 2 the volume susceptibility parallel to the trigonal axis of the crystal,

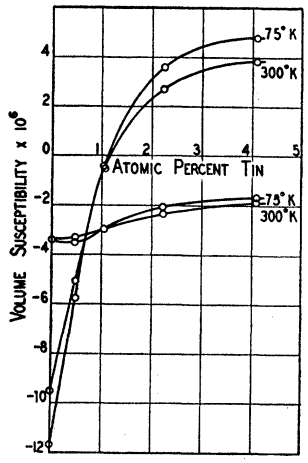

FIG. 4. Principal susceptibilities of Sb-Sn alloys.

$\kappa_{||}$ is plotted as a function of the absolute temperature, for each alloy used. Figure 3 shows similar graphs for the susceptibility perpendicular to the trigonal axis, $\kappa_{\perp}$. The behavior of $\kappa_{||}$ is of particular interest. It was observed that except for the crystal of 1.06 percent tin the diamagnetic susceptibilities increase toward diamagnetism and the paramagnetic susceptibilities increase toward paramagnetism at low temperatures. But when the observed values are corrected for the diamagnetism of the lattice ions, as explained in I, the origin of coordinates is shifted to $-1.32(10)^{-6}$, and all of the curves behave in the same way.

The susceptibilities $\kappa_{\perp}$ are only slightly dependent on the temperature. The curve for pure antimony shows complete temperature independence. No experimental points are indicated on this curve because of the method used to obtain it. The crystal was suspended with the proper orientation in the magnetic field. When liquid nitrogen was introduced, the reading of the microscope was closely observed while the system cooled to 77°K, and was taken at intervals during the rise back to room temperature. No apparent change in the reading was observed at any time.

The complete behavior of the antimony-tin alloys is shown by the curves of Fig. 4, in which the susceptibilities are plotted as a function of composition. The curves for 300°K are part of those given for these alloys in I. Points for the curves for 77°K were taken from Fig. 2 and Fig.

[8] R. L. Weber, *Temperature Measurement* (Edwards Brothers, Inc., Ann Arbor, Michigan, 1941), p. 163.

3. Points for intermediate temperatures would lie in the regions bounded by the curves shown. It is to be seen that when the origin of coordinates is shifted in accordance with the correction for the lattice ion susceptibility, the point of intersection of all the isothermals of $\kappa_{||}$ lies very nearly on the axis of abscissas, at approximately 0.93 atomic percent tin. This fact lends support to the estimate given in I for the overlap, and indicates that in this range of temperatures the

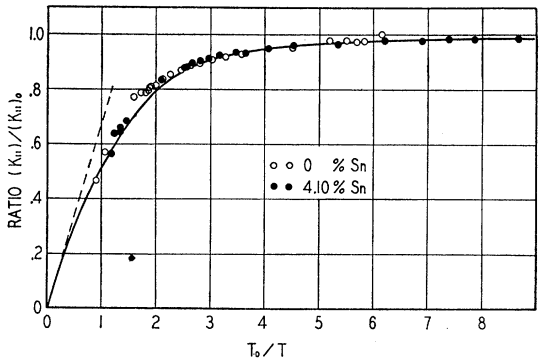

FIG. 5. Comparison of the magnetic susceptibilities of free electrons with $\kappa_{||}$, Sb-Sn alloys. Heavy line—theoretical curve.

overlap is essentially temperature independent.

The results of the measurements on pure antimony are in good agreement with those of Schoenberg and Uddin.[6] The values of the mass susceptibilities $\chi_{||}$ and $\chi_\perp$ at 90°K, from Fig. 2, are $-1.73(10)^{-6}$ and $-0.50(10)^{-6}$, respectively. Schoenberg and Uddin quote $\chi_{||}=-1.72(10)^{-6}$ and $\chi_\perp=-0.55(10)^{-6}$ at 90°K. They found $\chi_\perp$ to be independent of temperature.

DISCUSSION

The forms of the equations giving the susceptibilities of an electron gas, both for free and quasi-bound electrons, suggest that if the corrected susceptibilities be plotted against $1/T$, the resulting curves should approach a straight line through the origin for small values of $(1/T)\times(T\gg T_0{}^*$, where $T_0=\zeta_0/k)$, and should approach asymptotically some value $(\kappa_{||})_0$ for large values of $(1/T)(T\ll T_0{}^*)$. An experimental value of $T_0{}^*$, for each alloy, might then be found by extrapolating the curve to the origin and measuring the slope, as was done by Ganguli and Krishnan[9] for graphite. For the present case, however, this procedure was not very accurate, because measurements were made only below room temperature. To find $T_0{}^*$ and the best value of $(\kappa_{||})_0$, the experimental points were fitted to an equation of the form

$$\kappa_{||}=(\kappa_{||})_0\left\{1-\frac{\pi^2}{12}\left(\frac{T}{T_0{}^*}\right)^2\right\}$$

by the method of least squares. The fit was made for small values of T, in fact, for $T<180°$. The results obtained for all alloys are given in Table I.

If Eqs. (1) and (3) are divided by $\frac{3}{2}(n\mu^2/\zeta_0)$, and Eqs. (2) and (4) by $-\frac{1}{2}(n\mu^2/\zeta_0)$, the resulting expressions are identical for both the para- and diamagnetic susceptibilities. They are, for $kT\ll\zeta_0$,

$$1-\frac{\pi_2}{12}\left(\frac{T}{T_0}\right)^2,$$

and for $kT\gg\zeta_0$,

$$\frac{2}{3}\frac{T_0}{T}\left\{1-\frac{1}{3}\left(\frac{2}{\pi}\right)^{\frac{1}{2}}\left(\frac{T_0}{T}\right)^{\frac{3}{2}}\right\},$$

where the terms in H are neglected. If these functions are plotted against T_0/T, and joined together over the region where $T\sim T_0$, the curves shown in Fig. 5 and Fig. 6 result. To compare these curves with experiment, the corrected values of the susceptibilities, divided by $(\kappa_{||})_0$ are plotted against $T_0{}^*/T_0$ for each alloy. Figure 5 shows the points for 0 and 4.10 atomic percent tin, and Fig. 6 for the other three. The points in Fig. 5 which correspond to values of T_0/T below 1.5 were found for temperatures between 420°K and 530°K, to confirm the trend observed for lower temperatures. The agreement between the experimental values and the theoretical curve is good except in the region where $T\sim T_0{}^*$, as is to be expected.

TABLE I. *Susceptibilities of Sb-Sn alloys.*

At. % Sn	$(\kappa_{||})_0$	$T_0{}^*$ (°K)
0	$-10.60(10)^{-6}$	475
0.49	$-4.49(10)^{-6}$	585
1.06	$+0.92(10)^{-6}$	750
2.24	$5.01(10)^{-6}$	615
4.10	$6.18(10)^{-6}$	620

[9] N. Ganguli and K. S. Krishnan, Proc. Roy. Soc. **A177**, 168 (1941).

The value of T_0 for the overlapping electrons, if they were perfectly free, would be $T_0=1860°$K, from Eq. (6). Hence $\zeta_0^*=0.25\zeta_0$. This result suggests that, if the overlapping electrons are responsible for $\kappa_{||}$ the effect of the lattice field is to depress the upper limit of the Fermi distribution, that is, to increase the density of electronic states, but that otherwise they behave like free electrons.

From Eqs. (9) and (11) values of $\alpha_1\alpha_2$ and α_3 can be computed, provided n and ζ_0^* are known. Mott and Jones,[3] in discussing the magnetic properties of bismuth, have an estimate of the overlap, but no experimental information as to ζ_0^*. Using Eq. (11), and assuming $\alpha_3=1$, they evaluate the product $\alpha_1\alpha_2$, and hence ζ_0^*. A similar calculation for antimony yields $\alpha_1\alpha_2\sim 3(10)^3$, and $T_0^*\sim 15T_0=28{,}000°$. These assumptions lead to a value of κ_d/κ_p of the order of 10^3.

Ganguli and Krishnan, on the other hand, have available an experimental value of T_0^*. To evaluate the α's, they postulate the effective number of electrons as one per atom, and calculate T_0 on this basis. They then assume that $\alpha_1=\alpha_2$, and compute α_3. This procedure, for antimony, leads to $\alpha_3=(1/(88.6)^3)\sim 10^{-6}$. The assumption $\alpha_1=\alpha_2=1$ necessitates $\alpha_1\alpha_2=1$, so that the ratio $(\kappa_d/\kappa_p)=\frac{1}{3}$, as for free electrons, which means that the resulting susceptibility should be paramagnetic. This was not observed, in graphite, so Ganguli and Krishnan assume a spin-spin coupling to suppress the paramagnetism. This seems somewhat artificial.

The data available for antimony include both the number of overlapping electrons and the observed degeneracy temperature. If one uses these, and also the value $-10.6(10)^{-6}$ for κ_d in Eq. (9) he obtains $\alpha_1\alpha_2=52.5$. Hence $(\kappa_d/\kappa_p)=17.5$, a much more reasonable figure than 10^3, and one which makes the resultant susceptibility diamagnetic without the necessity of further assumptions.

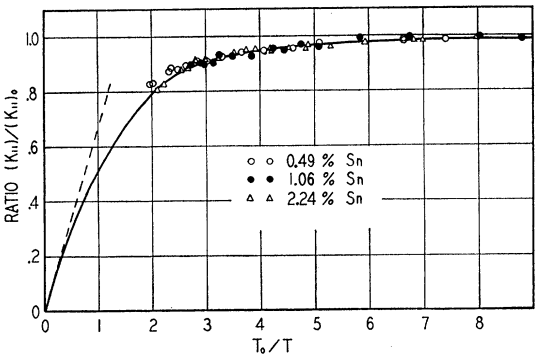

FIG. 6. Comparison of the magnetic susceptibilities of free electrons with $\kappa_{||}$, Sb-Sn alloys. Heavy line—theoretical curve.

As found above, if the overlapping electrons were prefectly free, they would have a $T_0=1860°$. Hence, from Eq. (12), $(\alpha_1\alpha_2\alpha_3)^{\frac{1}{3}}=0.25$, and so $\alpha_1=3.2(10)^{-4}$. These values of α_1, α_2, and α_3 in Eq. (7) represent an energy surface in **k** space somewhere between that of Mott and Jones on the one hand and Ganguli and Krishnan on the other.

Finally, having the value of T_0^*, the energy interval between the bottom of the second Brillouin zone and the top of the Fermi distribution is $kT_0^*=0.04$ ev for pure antimony.

Very little can be concluded from the experimentally observed behavior of $\kappa_\perp$, since the theory is not adequate to handle this case. The fact that $\kappa_\perp$ is so slightly dependent on temperature and on change in the electron concentration in the lattice suggests that it is in some way related to the electrons in the first Brillouin zone, but no quantitative information can be given.

One of us (S.H.B.) presented part of this work to the Faculty of the Graduate School of Yale University for the degree of Doctor of Philosophy. Part of the equipment used in this work was purchased by a grant from the George Sheffield Fund.