lute alloys spanning a wide range of  $0^{\circ}$ K susceptibilities indicates that the magnetic properties of alloys previously treated separately as spin-fluctuation and Kondo systems are qualitatively indistinguishable at sufficiently low temperatures. Moreover, the NMR results appear to be easily understood in terms of a single-particle description. This observation is consistent with the linear temperature dependence of the low-tempera<br>ture specific heats of Au:V<sup>19</sup> and Cu:Fe.<sup>20</sup> ture specific heats of  $Au:V^{19}$  and  $Cu:Fe.<sup>20</sup>$ 

It is a pleasure to acknowledge several helpful conversations with L. Dworin.

\*Work supported by U. S. Atomic Energy Commission.

 ${}^{2}$ N. Rivier and M. J. Zuckermann, Phys. Rev. Letters 21, 904 (1968).

 ${}^{3}$ N. Rivier, M. Sunjic, and M. J. Zuckermann, Phys. Letters 28A, 492 (1969).

 ${}^{4}$ H. Suhl, Phys. Rev. Letters 19, 442 (1967); M. J. Levine and H. Suhl, Phys. Rev. 171, <sup>567</sup> (1968); M. J. Levine, T. V. Ramakrishnan, and R. A. Wreiner, Phys. Rev. Letters 20, 1370 (1968).

5J. Kondo, Theoret. Phys. (Kyoto) 32, 37 (1964).

 $6A.$  D. Caplin, C. L. Foiles, and J. Penfold, J. Appl. Phys. 39, 842 (1968).

 ${}^{7}$ A preliminary account of this work appeared in A. Narath and H. T. Weaver, Bull. Am. Phys. Soc. 14, 371 (1969).

<sup>8</sup>The bulk properties have been discussed most recently by R. Aoki and T. Ohtsuka, J. Phys. Soc. Japan 26, 651 (1969).

<sup>9</sup>A. Narath and A. C. Gossard, Phys. Rev. (to be published).

 $^{10}$ K. Kume, J. Phys. Soc. Japan 23, 1226 (1967).

 $11$ M. D. Daybell, D. L. Kohlstedt, and W. A. Steyert, Solid State Commun. 5, 871 (1967).

 $^{12}$ A. Narath, K. C. Brog, and W. H. Jones, Jr., to be published.

 $13K$ . C. Brog, W. H. Jones, Jr., and J. G. Booth, J. Appl. Phys. 38, 1151 (1967).

 $^{14}$ K. C. Brog, W. H. Jones, Jr., and G. S. Knapp, Solid State Commun. 5, 913 (1967).

<sup>15</sup>Similar results for somewhat more concentrated Al:Mn alloys have been reported recently by Y. Oda, H. Yamagata, and K. Asayama, J. Phys. Soc. Japan 25, 629 (1968), whose shift value  $(-2.7\%)$  differs from ours, however. Our value is based on the  $55$ Mn nuclear moment determination of W. B. Mims, G. E. Devlin, S. Geschwind, and V. Jaccarino, Phys. Letters 24A, 481 (1967). Independent results for Al:Mn have also been obtained by H. Lanois and H. Alloul, Solid State Commun. 7, 525 (1969).

 $^{16}$ J. Butterworth, Proc. Phys. Soc. (London) 83, 71 (1964).

 $^{17}$ P. Lederer and D. L. Mills, Solid State Commun. 5, 131 (1967).

 $18$ Y. Yafet and V. Jaccarino, Phys. Rev. 133, A1630 (1964).

 $^{19}$ B. M. Boerstoel and W. M. Star, Phys. Letters 29A, 97 (1969); see also the earlier literature cited in Ref. 9.  $^{20}$ J. C. Brock, J. C. Ho, G. P. Schwartz, and N. E. Phillips, in Proceedings of the Eleventh International Conference on Low Temperature Physics, St. Andrews, Scotland, 1968, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrews University, St. Andrews, Scotland, 1968), p. 1229.

## INFLUENCE OF SMALL-ANGLE SCATTERING ON OPEN-ORBIT CONDUCTION IN THALLIUM\*

Robert E. Hamburg, f Claude G. Grenier, and Joseph M. Reynolds

Department of Physics and Astronomy, Louisiana State University and Agricultural and Mechanical College, Baton Bouge, Louisiana 70803

(Received 12 May 1969)

A measurement of the electrical and thermal magnetoresistance of thallium gives a direct demonstration of the scattering of open-orbit electrons into adjacent closed orbits through the mechanism of small-angle scattering. Potentially, this effect provides a method for determining the efficiency of small-angle scattering in metallic conduction.

In a metal, both electrical and thermal resistance depend greatly on how close an electron is brought to the equilibrium distribution after being scattered. Even though small-angle scattering would be efficient enough to cause thermal resistance, it is not generally expected to cause appreciable resistance to an electrical current. There are known exceptions, viz., the influence of small-angle scattering on the galvanomagnetie effects in metals has been discussed recently by

several authors.<sup>1</sup> Of these, Pippard's description of the expected behavior of the transverse magnetoresistance due to open-orbit electrons being scattered into adjacent closed orbits by small-angle scattering is the most appropriate for the work reported here.

In thallium a slice of open orbits exists on the honeycomblike fourth-zone electron sheet of the Fermi surface' when a magnetic field is applied close to the hexagonal direction. The narrow re-

<sup>&</sup>lt;sup>1</sup>P. W. Anderson, Phys. Rev. 124, 41 (1961).

gion of open orbits thus obtained is sandwiched between electronlike and holelike regions. In Fig. 1 such regions are schematized for a field applied at an angle  $\alpha$  with the [0001] direction in the  $(10\overline{1}0)$  plane. The shaded areas represent the closed-orbit regions and the open-orbit regions are represented by the nonshaded areas. The open orbits are extended in the  $\lfloor 10\overline{1}0 \rfloor$  direction with different orbit shapes occurring, depending on the position of the orbit within the slice. The width and thus the number of states in the openorbit region increases as the angle  $\alpha$  increases. This variation of slice width with increasing  $\alpha$ causes a change in the efficiency of the smallangle scattering, since at constant scattering angles electrons are more readily removed from the open orbit for narrow slices than they are for wider slices. It should be noted that when an electron is scattered from an open-orbit into a closed-orbit state its contribution to electrical conduction becomes negligible compared with its contribution in the open-orbit state. Moreover, as pointed out by Pippard, the inverse process, i.e., the scattering of electrons from closed to open orbits, does not compensate appreciably for this loss of conduction.

If  $r$  is the number of collisions necessary to scatter an electron from a point in the open-orbit region to a point in the adjacent closed-orbit region, then  $r$  will depend on the initial position of the electron in the open-orbit slice, the width of the slice, and the type of scattering involved. For example, in the case of small-angle scattering due only to low-energy phonons, the number of collisions  $r<sub>p</sub>$  depends on the phonon distribution. Therefore, averaging the initial states of the electrons over the width of the open-orbit region, the average numbers of collisions  $\bar{r}$  and  $\bar{r}_p$ necessary for electron relaxation are expected to depend on the width of the slice and the phonon distribution, i.e., the tilt angle  $\alpha$  and the temperature.

If both open and closed orbits exist simultaneously in a metal, the total conductivity tensor can be written as

$$
\hat{\sigma}^T = \hat{\sigma}^C + \hat{\sigma}',
$$

where  $\hat{\sigma}^c$  is the contribution to the conductivity from the closed orbits and  $\hat{\sigma}'$  is the contribution due to the open orbits. When  $\alpha$  is small, the closed-orbit carriers still correspond to the case of a two-dimensional isotropic metal with a con-



FIG. 1. Schematic representation of the band of open orbits on the fourth electron zone in thallium as it will appear when the magnetic field is tilted by a small angle  $\alpha$  from the hexagonal direction toward the [1210] direction, represented as the nonshaded area. The open orbits extend in the  $[10\bar{1}0]$  direction in k space. The main shaded area corresponds to the adjacent closed hole orbits. The smaller shaded areas correspond to adjacent closed electron orbits. The main open-orbit band is also adjacent to paraIlel open-orbit regions from neighboring cells, but the only smallangle scattering which is efficient displaces an electron from the open-orbit region to an adjacent closed-orbit region.

ductivity

$$
\hat{\sigma}^c = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} \\ -\sigma_{xy} & \sigma_{xx} \end{pmatrix},
$$

and for the open orbits,

$$
\hat{\sigma}' = \begin{pmatrix} \sigma' \cos^2\theta & \sigma' \cos\theta \sin\theta \\ \sigma' \cos\theta \sin\theta & \sigma' \sin^2\theta \end{pmatrix},
$$

where  $\theta$  is the angle between the real-space openorbit direction and the current direction.

The total resistivity  $\widehat{\rho}^{\,T}$  is given by  $\widehat{\rho}^{\,T}$  = ( $\sigma^{\,T}\!)^{-1}$ ; so by inverting the conductivity matrix and taking the known high-field limit for the components of the closed orbit conductivity, viz.,  $\sigma_{xx} \rightarrow A/H^2$ 

and  $\sigma_{xy} \rightarrow B/H$ , then the high-field transverse magnetoresistance is

$$
\rho_{xx} \approx \frac{A}{B^2} \frac{1 + (H^2 \sigma'/A) \sin^2 \theta}{1 + \sigma' A/B^2}.
$$

For small tilt angles  $\alpha$ , it is expected and a posteriori verified that

$$
\sigma' A/B^2 \ll 1;
$$

so

$$
\rho_{xx} \approx A/B^2 + (\sigma'/B^2)H^2 \sin^2\theta. \tag{1}
$$

Neglecting the lattice conductivity  $\lambda_{g}$ , the corresponding equation for the electronic part of the high- field thermal magnetoresistance is

$$
\gamma_{xx} \approx A'/B'^2 + (\lambda'/B'^2)H^2 \sin^2\theta, \qquad (2)
$$

where  $\lambda'$  is the open-orbit thermal conductivity, and  $A'$  and  $B'$  are defined by the high-field approximations

$$
\lambda_{rr} \approx A'/H^2
$$

and

 $\lambda_{xy} \approx B'/H$ 

A plot of  $\rho_{xx}$  vs  $H^2$  yields a straight line whose slope is  $\sigma' B^{-2} \sin^2 \theta$ . By scaling  $\gamma_{xx}$  with the ideal Wiedemann-Franz ratio  $L_nT$ , a plot of  $L_n T \gamma_{xx}$  vs  $H^2$  yields a straight line whose slope is  $L_n T \lambda' B'^{-2} \sin^2 \theta$ . Since  $B' = BL_nT$ , then the ratio of the two slopes is  $\overline{r} = \sigma'L_n T/\lambda'$ , and with  $\sigma'$ and  $\lambda'/L_pT$  being, respectively, proportional to  $\tau_{\alpha}$  and  $\tau_{\lambda}$ , their corresponding times of relaxation,  $\vec{r}$  also gives the ratio  $\tau_o / \tau_{\lambda}$ . One expects that  $\tau_{\lambda}$  should give a good average value of the time between collisions of all kinds  $\tau_c$ ; so  $\bar{r}$  as previously defined gives the average number of times an open-orbit electron is scattered before reaching the equilibrium distribution for the electrical process.

A single crystal of thallium was cut in the shape of a rectangular slab and mounted with the long side vertical. This vertical or  $x$  direction was in the [10TO] crystallographic direction and was the direction in which the heat or electrical current was applied. The hexagonal direction [0001]was horizontal and perpendicular to the large face of the sample so that the magnetic field direction (z direction) could be rotated to form the angle  $\alpha$  with the hexagonal direction. The general direction of the open orbit in  $k$  space was then along  $[10\overline{1}0]$  and the open orbits in real space were perpendicular to the current direction  $(x$  direction). Thus, should the field direction to crystal-orientation conditions exactly meet, one expects  $|\theta| = 90^{\circ}$  except for  $\alpha = 0$ . Electrical- and thermal-resistivity data were taken as a function of temperature as well as a function of the magnitude and direction of the magnetic field.

A plot of  $\overline{r} = \tau_o/\tau_{\lambda}$  vs  $\alpha$  is shown in Fig. 2. This set of curves shows clearly that for all temperatures  $\bar{r}$  decreases as expected when  $\alpha \rightarrow 0$ , i.e., as the open-orbit slice width decreases. It also shows that at higher temperatures the decrease appears for greater slice widths. This is because at higher temperatures the average phonon wavelength  $\bar{q}$  is larger  $(\bar{q} \propto T)$  and the same number of scatterings displaces an electron farther on the Fermi surface, i.e., out of a wider slice of the open orbits. Thus, the width of the dip in the  $\bar{r}$  curve depends directly on the magnitude<sup>3</sup> of  $\bar{q}$ . At 4.1°K, for example,  $\bar{q} \approx 14 \times 10^6$ 



FIG. 2. The ratio  $\overline{r}$  between the open-orbit conductivities  $\sigma'$  and  $\lambda'/L_nT$  gives an approximate measurement of  $\tau_{\alpha}/\tau_{\lambda}$  for the open orbits, i.e., the approximate number of all types of collisions necessary to relax an electron in the electrical conduction. This number decreases when the open-orbit zone width is decreased making small-angle scattering efficient in removing the electron from the open-orbit zone into a closed-orbit zone. The experimental points and error flags are shown only for the  $4.1^{\circ}$ K data for better clarity. The left side shows the effect for  $\alpha$  positive. The  $+\alpha$  and  $-\alpha$ . These data are not corrected for  $\lambda_g$ . right side of the figure shows a corrected average for

 $cm^{-1}$ . This value corresponds to the averaged slice width which would result from a tilt angle of  $\alpha \approx 2.5^\circ$ . The few-degree range over which the dip in the  $\bar{r}$  curve extends appears, therefore, to be correct.

One would expect  $\bar{r}$  to tend toward unity as  $\alpha$  $\rightarrow$  0. However, as  $\sigma' \sin^2 \theta$  tends toward zero, the corresponding thermal term tends toward  $\lambda_{\alpha}$ , the neglected lattice conductivity, which at  $4.1^{\circ}$ K may be large enough to be detectable when the electronic part becomes vanishingly small. Thus the apparent decrease of  $\bar{r}$  below unity at 4.1°K is to be expected. The empirical corrections attempted to correct for the existence of  $\lambda_g$  show that the dip in the  $\bar{r}$  curve is primarily due to the effect of small-angle scattering even though  $\lambda_{\alpha}$  affects the shape of this curve.

At the larger slice widths, where the bulk-type conditions prevail, the impurity scattering (bound ary scattering included) is seen to cause smaller  $\bar{r}$  values at the lowest temperature. In order to account for this, the data might be reanalyzed by writing

$$
\overline{r} = \frac{\tau_{\lambda p}^{-1} + \tau_{\text{imp}}^{-1}}{\tau_{op}^{-1} + \tau_{\text{imp}}^{-1}},
$$

where the index  $p$  refers to the phonon-scattering contribution only, and "imp" refers to the contribution due to impurities. A preliminary determination of the quantity  $\bar{r}_p = \tau_{op}/\tau_{\lambda p}$  was attempted together with the empirical correction for  $\lambda_g$ . Curves similar to the  $\bar{r}$  curves of Fig. 2 were drawn for  $\bar{r}_p$  so that an analysis of the pure small-angle scattering could be attempted. However, the experimental errors in the existing data and the indetermination of  $\lambda_g$  made the result of this analysis inconclusive.

Improvements in the experimental apparatus and procedure are expected to reduce some of the errors and yield more accurate values for  $\sigma'$ ,  $\lambda'$ ,  $\bar{r}$ , and  $\bar{r}_p$  from which quantitative conclusions about electron-phonon small-angle scattering may then be drawn.

The small-angle-scattering mechanism studied here is expected to occur in any metal where open orbits are adjacent to closed orbits. This situation occurs in a large number of metals, and the present method of studying this effect is, in principle, applicable with only slight modifications for most cases. $4$ 

uscript is gratefully acknowledged. Present address: Department of Physics, Louisiana Polytechnic Institute, Ruston, La.

'Some general considerations on small-angle scattering can be found, for example, in the following articles: A. B. Pippard, Proc. Roy. Soc. (London), Ser. <sup>A</sup> 305, <sup>291</sup> (1968); P. G. Klemens and J. L. Jackson, Physica 30, 2031 (1964), and 31, 1421 (1965). Effects of small-angle scattering for the case of specific metals are discussed by P. Cotti, E. M. Fryer, and J. L. Olsen, Helv. Phys. Acta 37, 585 (1964); Robert L. Powell, in Proceedings of the Ninth International Conference on Low Temperature Physics, edited by J. G. Daunt, D. O. Edwards, F.J. Milford, and M. Yaqub (Plenum Press, Inc., New York, 1965), Pt. B, p. 732; J. O. Ström-Olsen, Proc. Roy. Soc. (London), Ser. A 302, 83 (1967}. Localized umklapp scattering has been studied by H. A. Young, Phys. Hev. 175, 813 (1968}, and intersheet scattering by O. P. Eatyal, A. N. Gerritsen, J. Buvalds, H. A, Young, and L. M. Falicov, Phys. Hev. Letters 21, <sup>694</sup> (1968); R. A. Young, J. Huvalds, and L. M. Falicov, to be published. References to earlier work may be found in the articies listed above.

 ${}^{2}$ A relativistic orthogonalized-plane-wave calculation of the band structure and fs of thallium made by P. Soven, Phys. Rev. 137, A1706, A1717 (1965) is in relatively good agreement with experimental determinations of the thallium fs by the de Haas-Van Alphen effect: M. G. Priestley, Phys. Rev. 148, 580 (1966), and by magnetoacoustic attenuation: J. A. Rayne, Phys. Rev. 131, <sup>653</sup> (1963); Y. Eckstein, J.B.Ketterson, and H. G. Priestley, Phys. Rev. 148, 586 (1966); and J.B.Coon, C. G. Grenier, and J. M. Reynolds, J. Phys. Chem. Solids 28, 301 (1967). For a study on open orbits in thallium see J. C. Milliken and R. C. Young, Phys. Rev. 148, 558 (1966), where reference to previous work is given.

 $^3\! \bar{q}$  may be taken  $\approx$  2.6 $q$ <sub>D</sub>T/ $\Theta$ <sub>D</sub>, viz., the value for which the electron-phonon scattering is a maximum, i.e., where  $x^3 e^{x}/(e^x-1)^2$  is a maximum, with  $x = (q/q_{\text{D}})^2$  $(T/\Theta_{\rm D})^{-1}$ ,  $q_{\rm D}$  and  $\Theta_{\rm D}$ , being the Debye sphere radius and temperature, respectively.

<sup>4</sup>A review of the effect of open orbits on magnetoresistance can be found in E. Fawcett, Advan. Phys. 13, 139 (1964), in which references, theory, and a partial list of open-orbit metals is given. The open-orbit  $H^2$ dependence in thallium is characteristic of open orbits of "type II" (or "aperiodic") corresponding to "twodimensional regions" on the open-orbit stereograms. <sup>A</sup> similarity with thallium exists if the open-orbit "region" surrounds a three-, four-, or sixfold symmetry direction as in Cu, Ag, Au, Sn, Nb, Ta, etc., and only a slight modification of the method used in Tl is necessary to include the case where the "region" corresponds to a twofold axis as in Pb, Ag, etc. The possibility of extending the method to "type I" (or "periodic") open orbits corresponding to a "one-dimensional region" would depend on the particular metal involved. The useful range of temperatures where study should be carried out would be in a direct relation with the Debye temperature of the metal.

<sup>\*</sup>Work performed under the auspices of the U. S. Atomic Energy Commission, and U. S. Atomic Energy Report No. ORO-3087-33.

<sup>/</sup>Financial assistance provided by the Dr. Charles E. Coates Memorial Fund for the preparation of this man-