man, and Professor H. Ehrenreich for many stimulating discussions. The technical assistance of R. Dykeman is gratefully acknowledged.

<sup>1</sup>R. W. Smith, Phys. Rev. Letters <u>9</u>, 87 (1962). <sup>2</sup>J. H. McFee, J. Appl. Phys. 34, 1548 (1963). <sup>3</sup>H. Kroger, E. W. Prohofsky, and R. W. Damon, Phys. Rev. Letters 11, 246 (1963).

<sup>4</sup>E. W. Prohofsky, Phys. Rev. (to be published). <sup>5</sup>L. Spitzer, Jr., <u>Physics of Fully Ionized Gases</u> (Interscience Publishers, Inc., New York, 1956), pp. 58-64.

<sup>6</sup>J. B. Gunn, Solid State Commun. 1, 88 (1963).

## MAGNETORESISTANCE AND MAGNETIC BREAKDOWN\*

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The galvanomagnetic properties of metals have been widely used to study the motion of the conduction electrons.<sup>1</sup> Generally, for high magnetic fields, two distinct types of transverse magnetoresistance  $\rho_{\perp}(H)$  are found, either a resistance that saturates, i.e., approaches a constant value as the field is increased, or a resistance which increases without bound as  $H^2$ . These properties can be uniquely related to the presence or absence of open trajectories.<sup>2</sup> The appearance of quadratic behavior indicates (a) in metals with an odd number of conduction electrons per unit cell: the existence of open trajectories with an average direction in k space not perpendicular to the direction of the current; (b) in metals with an even number of conduction electrons per unit cell: either no open orbits and equal "volume" of electron and hole trajectories (the so-called compensated case), or open orbits not perpendicular to the current.

When high magnetic fields are used in metals which have relatively small energy gaps, the dynamics of the motion is such that there is a finite probability for the electrons to ignore such gaps and describe trajectories which connect two different pieces of Fermi surface (thus changing the character of the orbits). This effect is called magnetic breakdown,<sup>3-5</sup> and it has been found experimentally in several metals, mostly those with hexagonal close-packed structure.<sup>6-10</sup> The probability of breakdown, that is, the probability for an electron to make a transition between two different orbits, has been found by Blount<sup>4</sup> to be

$$P = \exp[-(H_0/H)], \qquad (1)$$

$$H_0 = K \Delta^2 m c / E_F e, \qquad (2)$$

where  $\Delta$  is the energy gap,  $E_{\mathbf{F}}$  the Fermi energy, and K a constant of the order unity.<sup>11</sup>

When orbits of different character are coupled by magnetic breakdown, new types of behavior must be expected for  $\rho_{\perp}(H)$ . In particular, when the two limiting cases  $P \rightarrow 0$  and  $P \rightarrow 1$  correspond to nonsaturation and saturation, respectively,  $\rho_{\perp}(H)$  is likely to exhibit a maximum at fields of the order of  $H_0$ .

We have computed <u>semiclassically</u> the transverse conductivity and resistivity tensors by means of a modification of Chambers' pathintegral method,<sup>12</sup> which includes the possibility of magnetic breakdown at a finite number of points in the orbit. This new method involves a matrix M, a function of P only, which connects in the proper way the path integrals over pieces of the Fermi surface on which the electron trajectories develop. This results in a <u>classical</u> network, similar to the <u>quantum-mechanical</u> network introduced by Pippard<sup>5,13</sup> in the study of the quantization of coupled orbits.

We have solved a large number of cases, all two-dimensional (i.e., neglecting longitudinal effects), which result from a spherical Fermi surface with a finite number of Bragg reflections. The intrinsic relaxation time was assumed to be constant throughout. We included cases with no axis of symmetry and with two-, four-, and sixfold symmetries. We also studied magnetic breakdown which corresponded to transitions between the following kinds of orbits: (I) open + electron  $\rightarrow$  electron, (II) open + electron + hole  $\rightarrow$  electron + hole (compensated), (III) hole - electron, (IV) electron + hole (compensated)  $\rightarrow$  electron + electron. An example of class (I) is shown in Fig. 1, where transitions between an "undulating cylinder" and a "lens" make the final orbit equal to the original circle. Figure 1(c) shows the transverse magnetoresistance in the x direction as a function of  $\omega \tau$ , where  $\omega = eH/mc$  is the cyclo-



FIG. 1. Magnetic breakdown between open orbits and closed orbits. (a) The orbits in k space at P = 0; (b) the orbits in k space at P = 1; (c) magnetoresistance  $\rho_{11}$  in the x direction as a function of  $\omega \tau$  for various values of breakdown field  $\omega_0 \tau$ .  $\rho_{11}$  is in arbitrary units.

tron frequency, and as a function of the parameter  $\omega_0 \tau$ , where  $\omega_0 = eH_0/mc$  and  $H_0$  is defined in (1) and (2). Similarly, Fig. 2 shows an example of class (IV) with sixfold symmetry.

From these calculations the following conclusion can be drawn:

(i) Cases (I) and (IV) show an initial increase of magnetoresistance as  $\omega^2$ , a maximum value at  $\omega_m$ , and a decrease towards a saturation value  $\rho_{sat}$ , different from the initial  $\rho_0 = \rho(H = 0)$ .

(ii)  $\omega_m$  is only a fraction of  $\omega_0$ , of the order of  $\frac{1}{3}$  to  $\frac{1}{10}$ , and it moves to the lower values as the purity of the sample is increased. For instance, for the case shown in Fig. 2,  $\omega_m$  satisfies approximately the equation

$$\omega_0 \cong 3.14 \omega_m + 0.0356 \omega_m^2 \tau.$$
 (3)

(iii) The saturation value of the magnetoresistance,  $\rho_{\rm Sat},$  in cases (I) and (IV) satisfied the equation

$$\rho_{\text{sat}} = \rho_0 (1 + C \omega_0 \tau), \qquad (4)$$

where C is a constant of the order unity. In the cases of Figs. 1 and 2, C takes the values  $4/\pi$  and  $\sqrt{3}/2$ , respectively. If we take into account the fact that  $\rho_0 \propto \tau^{-1}$ ,

$$\rho \propto (\tau^{-1} + C \omega_0), \qquad (5)$$

and consequently it should approach a constant value as  $\omega_0 \tau \gg 1$ . Equations (4) and (5) can be easily interpreted by assuming a change in the effective relaxation time  $\tau_{eff}$  due to an additional scattering mechanism associated with incoherent Bragg reflection after breakdown is complete and such that

$$\tau_{\rm eff}^{-1} = \tau^{-1} + C\omega_0.$$
 (6)

(iv) The influence of more complicated orbits, possible only in the case of intermediate probabilities P, can be partly analyzed by looking at the off-diagonal components  $\rho_{12}$ ,  $\rho_{21}$  (Hall resistance) of the magnetoresistance tensor. From Fig. 2(d) it is possible to see that initially the "effective" number of electrons and holes is



FIG. 2. Magnetic breakdown between compensated (electron-plus-hole) orbits and uncompensated (electron) orbits. (a) The broken-down electron orbit at P = 1; (b) the hole orbit at P = 0: (c) the transverse magnetoresistance  $\rho_{11} = \rho_{22}$ ; (d) the Hall resistance  $\rho_{12} = -\rho_{21}$ . All values  $\rho$  in arbitrary units.

equal; the presence of extended hole orbits at intermediate ranges makes the "effective" number of holes larger than the number of electrons, while at higher fields the orbits reduce all to electron type, approaching asymptotically to the P=1 curve. It is interesting to note that the change in sign in Fig. 2(d) takes place at values of  $\omega$  close to  $\omega_0$ .

(v) Complete analysis of the other cases studied has been carried on in a similar fashion and with similar results.

It should be noted that the case of Fig. 2 corresponds fairly closely to the actual cases found in Mg<sup>6,14</sup> and Zn<sup>8,14</sup> for magnetic fields parallel to the hexad axis. In fact, curves very similar to Fig. 2(c) have been observed by Stark<sup>14</sup> for both Mg and Zn. The general features described above seem to agree well with experiment. Oscillatory behavior, small in Mg but very large in Zn, corresponds to quantum effects not included in our semiclassical approach. The oscillations are due to the Landau levels discussed by Pippard.<sup>5,13</sup> Their effect on the galvanomagnetic properties at high fields could possibly be included by assuming an oscillatory probability P similar to that proposed by Stark,<sup>8</sup> particularly when the second term in (6) dominates. In this way no problem of gauge invariance arises.

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<sup>&</sup>lt;sup>1</sup>See, for instance, R. G. Chambers, in <u>The Fermi</u> <u>Surface</u>, edited by W. A. Harrison and M. B. Webb (John Wiley & Sons, Inc., New York, 1960), p. 100; and the many references quoted there.

<sup>&</sup>lt;sup>2</sup>I. M. Lifschitz, M. Ya Azbel', and M. I. Kaganov, Zh. Eksperim. i Teor. Fiz. <u>30</u>, 220 (1955) [translation: Soviet Phys.-JETP <u>3</u>, <u>143</u> (1956)]; <u>31</u>, <u>63</u> (1956) [translation: Soviet Phys.-JETP <u>4</u>, <u>41</u> (1957)]. I. M. Lifshitz and V. G. Peschanskii, Zh. Eksperim. i Teor.

Fiz. 35, 1251 (1958) [translation: Soviet Phys.-JETP 8, 875 (1959)]; 38, 188 (1960) [translation: Soviet Phys. - JETP 11, 137 (1960)].

<sup>3</sup>M. H. Cohen and L. M. Falicov, Phys. Rev. Letters <u>7</u>, 231 (1961). <sup>4</sup>E. I. Blount, Phys. Rev. <u>126</u>, 1636 (1962).

<sup>5</sup>A. B. Pippard, Proc. Roy. Soc. (London) A270, 1

(1962); and (to be published). <sup>6</sup>M. G. Priestley, L. M. Falicov, and G. Weisz,

Phys. Rev. 131, 617 (1963).

<sup>7</sup>R. W. Stark, T. G. Eck, W. L. Gordon, and

F. Moazed, Phys. Rev. Letters 8, 360 (1962).

<sup>8</sup>R. W. Stark, Phys. Rev. Letters 9, 482 (1962).

<sup>9</sup>A. R. Mackintosh, L. E. Spanel, and R. C. Young, Phys. Rev. Letters 10, 434 (1963).

<sup>10</sup>A. S. Joseph and A. C. Thorsen, Phys. Rev. Letters 11, 67 (1963).

<sup>11</sup>Blount has derived Eq. (1) only for the limits of small or large H, but it is reasonable to assume that the expression is approximately valid for all fields.

<sup>12</sup>R. G. Chambers, Proc. Phys. Soc. (London) A65, 458 (1952).

<sup>13</sup>A. B. Pippard (private communication and to be published).

<sup>14</sup>R. W. Stark (private communication and to be published).

## LONG-RANGED MAGNETIC POLARIZATION EFFECTS

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Two years ago a group<sup>1</sup> at Bell Telephone Laboratories discovered an exceedingly long-ranged magnetic interaction among rare earth (RE) atoms dissolved in palladium. It is the purpose of the present note to suggest a theory for this phenomenon, which, it will be recalled, could not be explained by the Ruderman-Kittel interaction

$$J_{ij}^{\mathbf{R}-\mathbf{K}} = A(\sin x - x \cos x)/x^4, \quad x = 2k_{\mathbf{F}}R_{ij}, \quad (1)$$

nor even by the longer ranged Yosida modification,

$$J_{ij}^{Y} = A'(\sin x - x \cos x)/x^{3}, \qquad (2)$$

without assuming an unreasonably small value of  $k_{\rm F}$ . In fact, the experimenters had to adopt an ad hoc interaction<sup>1</sup>

$$J_{ij}^{\text{BTL}} = A'', \quad R_{ij} < r_0,$$
  
= 0,  $R_{ij} > r_0,$  (3)

such that any two RE atoms separated up to a distance  $r_0$  (~15 Å) underwent an interaction A''. No physical explanation was proposed for this novel interaction, nor do we know of any published to date.

We propose the possibility that the physical mechanism is none other than the usual indirect exchange via conduction electrons, but modified to include exchange matrix elements which change the total angular momentum of the f shell by  $\pm \hbar$ . and suggest Eq. (10) to replace Eq. (3). It has

already been pointed out by Brout and Suhl<sup>2</sup> that such processes add a gap (in excess of 0.1 eV) to the Ruderman-Kittel energy denominators. Consider the complete indirect exchange Hamiltonian

$$H = -\sum J(R_{ij})\vec{J}_i \cdot \vec{J}_j, \qquad (4)$$

where

$$\begin{split} J(\boldsymbol{R}_{ij}) &= B \sum_{f_k} (1 - f_{k'}) \exp[i(\vec{\mathbf{k}} - \vec{\mathbf{k}'}) \cdot \vec{\mathbf{R}}_{ij}] \\ &\times \left( (g_i - 1)(g_j - 1)(E_{k'} - E_k)^{-1} + \exp(i\pi\eta_j) \right) \\ &\times \left\{ (g_i - 1) + \exp(i\pi\eta_i) [S_i(S_i + 1) - J_i(J_i + 1)(g_i - 1)^2]^{1/2} \right\} \\ &\times [S_j(S_j + 1) - J_j(J_j + 1)(g_j - 1)^2]^{1/2} (E_{k'} + \Delta - E_k)^{-1} \right). \end{split}$$
(5)

The quantity  $(g_i - 1)$  is like a "charge": negative for the rare earths to the left of Gd in the periodic table, and positive for those to the right. The sign factor  $\eta_i = 0$  or 1 depends on the relative phases of the ground state  $(J_i)$  and the excited state  $(J_i \pm 1)$ . Both terms in Eq. (5) contribute to the Ruderman-Kittel interaction, although the contributions may add or cancel depending on the relative sign. In addition, the second term has a monotonic long-ranged part that we isolate by means of the identity.

$$(E_{k'} + \Delta - E_{k})^{-1} = (E_{k'} - E_{k})^{-1}$$
$$- \Delta (E_{k'} - E_{k})^{-1} (E_{k'} + \Delta - E_{k})^{-1}, \qquad (6)$$