Logarithmic dependence of the low-field magnetoresistance in $Hg_{3-\delta}AsF_6$

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(Received 16 September 1980)

A Fermi surface with a square cross section and slightly rounded corners, which results from perpendicular sets of weakly coupled one-dimensional chains, is found both experimentally and theoretically to lead to a strong enhancement of the low-field magnetoresistance, and a logarithmic dependence on the magnetic field, $\Delta\rho(H)/\rho(0) \propto H^2 \ln H_0/H$ for $H < H_0$, where H_0 is a characteristic effective field. The logarithmic dependence is observed for $Hg_{3-\delta}AsF_6$ in a series of new high-sensitivity experimental results for $0.5 < H < 40$ Oe at a temperature of 4.2 K. Analysis of the data leads to a value for the weak interchain coupling ($\Delta E/E_F \simeq 6 \times 10^{-3}$). Hall-effect data are presented for the temperature range $4.2 < T < 100$ K, and fields up to 4.5 kOe. At low temperatures, the measured Hall coefficient is in agreement with the anisotropic three-dimensional Fermi surface determined earlier by de Haas–van Alphen measurements. The smaller Hall coefficient above 10 K suggests the possibility of a crossover to localization onto families of parallel one-dimensional chains.

I. INTRODUCTION

The unusual results of electrical transport studies of the linear chain conductor $Hg_{3-\delta}AsF_6$ have been of considerable interest.[1–3] In spite of the presence of a high concentration of structural defects[4] in a quasi-one-dimensional structure,[5] the resistivity is strongly temperature dependent, decreasing monotonically with decreasing temperature in a metallic fashion, with no indication of residual resistivity even at the lowest temperatures. The enhanced magnetoresistance[2,3] observed in $Hg_{3-\delta}AsF_6$ has been attributed to the unusual Fermi surface which results from the perpendicular sets of relatively weakly coupled linear Hg chains.[4,5] In particular, low-field studies[2] showed that $\Delta\rho(H)/\rho(0)$ approached quadratic behavior only for $H \sim 1$ G. In this paper we present galvanomagnetic data; more extensive and accurate low-field-magnetoresistance data are reported, the Hall-effect results are reported here for the first time. We show that the magnetoresistance at low magnetic fields varies as $\Delta\rho(H)/\rho(0) \propto H^2 \ln H_0/H$ where H_0 is a characteristic effective field (we find $H_0 \simeq 130$ Oe). The $\ln H$ factor results directly from the structure of the cylindrical Fermi surface with a nearly square cross section.[6] New high sensitivity experimental results are presented in the range of $0.5 < H < 40$ G which confirm the logarithmic dependence. The low-temperature Hall-effect results are consistent with the Fermi-surface model of Razavi *et al.*[6] at low temperatures. On the other hand, Hall effect data above 10 K suggest a three-dimensional (3D) to one-dimensional (1D) crossover in electronic structure.

II. EXPERIMENTAL TECHNIQUES AND RESULTS

A. Low-field magnetoresistance

In our previous experimental studies,[2] we noted that even at low fields ($H < 10$ G), $\Delta\rho(H)/\rho(0)H^2$ was not constant. In order to determine the field dependence with more accuracy we have carried out additional low-field magnetoresistance studies at 4.2 K using a high-sensitivity contactless mutual inductance apparatus designed to allow measurements to the lowest magnetic fields ($H < 1$ G).

A schematic diagram of the electronics and Hartshorne bridge circuitry is shown in Fig. 1. The electrical conductivity is determined by the off-balance signal of the bridge, induced upon inserting the sample into one of equivalent secondary (S) coils. In this experiment, in order to extend the measured magnetoresistance to the lowest possible fields, it was necessary to separate the small field-dependent change in the conductance of the sample from the large field-independent background. Before applying the dc magnetic field, the bridge was balanced with the sample in the secondary coil $S1$ and a copper ring in $S2$. The shape and size of the copper ring were modified until a good balance of both the real and imaginary parts of V_{out} were achieved with the apparatus at 4.2 K. The magnetoresistance of the copper balance ring was measured separately at high magnetic fields. The results imply a change of less than 10^{-7} in 1 G. Thus the output of the balanced system was due entirely to the magnetoresistance of the single-crystal $Hg_{3-\delta}AsF_6$ samples. The signal-to-

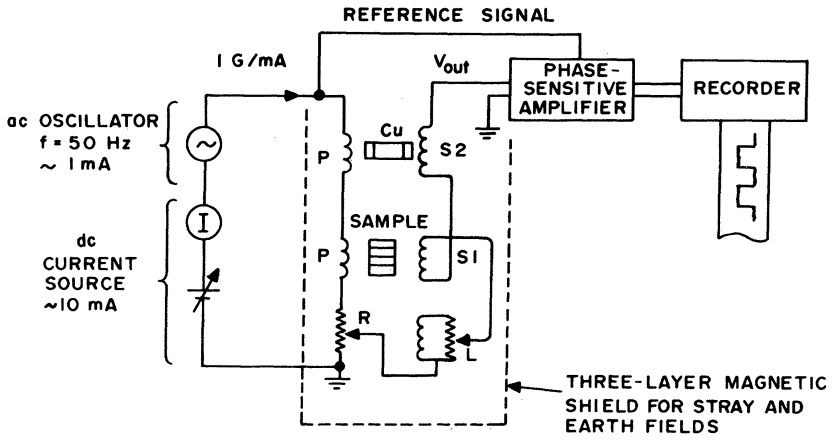

FIG. 1. Schematic diagram of the electronics and Hartshorne bridge. P are the primary coils, $S1$ and $S2$ are the secondary coils, and L and R are for adjusting the complex impedance to obtain balance ($V_{out}=0$).

noise ratio was such that the minimum detectable change was $\Delta\rho(H)/\rho(0) \simeq 5\times10^{-6}$.

The main source of short as well as long term noise was thermal fluctuations of the ^{4}He bath. Temperature stability is especially important in this case since the low-temperature resistivity of $Hg_{3-\delta}AsF_6$ varies approximately at T^3.[2,7,8] Consequently, thermal fluctuations of order 10^{-4} K set a value of about 0.5 G as the lower limit on the dc magnetic field.

The experimental results are shown in Fig. 2 where we plot $\Delta\rho(H)/\rho(0)H^2$ vs $\ln H$ at 4.2 K for several values of the ac driving field (h_{ac}). For comparison, the four sets of data were normalized (at $H=8$ G) to the data from $h_{ac}=0.86$ G. The results shown in Fig. 2 confirm the unusual field dependence found earlier[2] and demonstrate that the functional dependence of $\Delta\rho(H)/\rho(0)$ is $H^2|\ln H|$ above 2 G, saturating to quadratic behavior at lower fields.

B. Hall effect

Hall-effect measurements were carried out using the generalized Montgomery method[9] developed by Spal.[7]

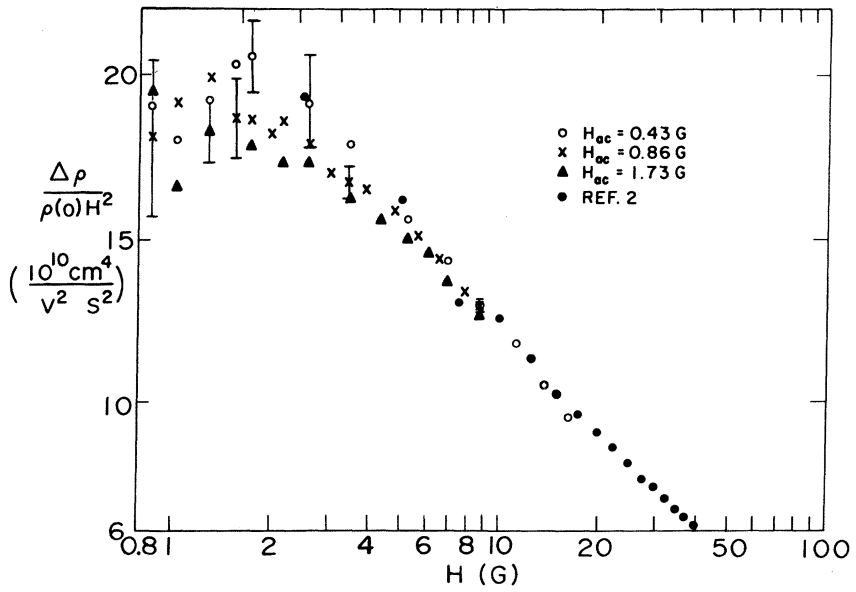

FIG. 2. $\Delta\rho(H)/\rho(0)H^2$ at 4.2 K as a function of $\ln H$. The logarithmic dependence [Eq. (9)] is evident above 2 G.

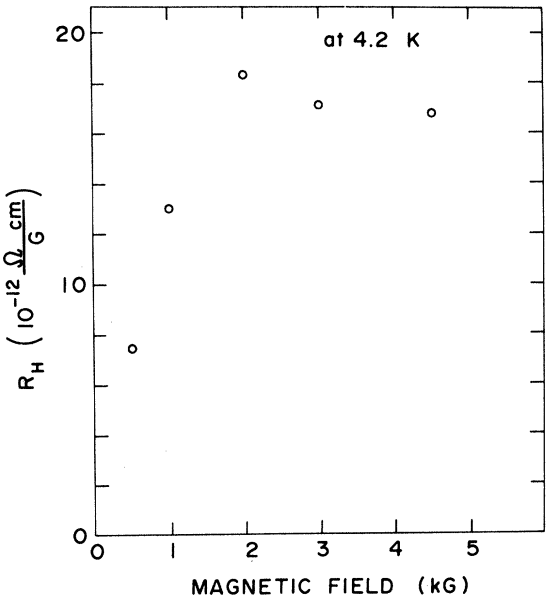

FIG. 3. Hall coefficient (R_H) vs magnetic field at 4.2 K.

A well-faceted single crystal was cut with dimensions $8.9 \times 8.9 \times 1.1$ mm^3; the top and bottom planes of the samples were coincident with the crystallographic *ab* plane. All sample handling and mounting were done in a high-quality controlled-atmosphere dry box filled with argon. After mounting, the sample assembly was sealed into a can containing helium exchange gas. The dc measurements covered the magnetic field range $H < 5$ kG.

The *ab* plane Hall coefficient, R_H, of $Hg_{3-\delta}AsF_6$ was measured as a function of magnetic field at 4.2 K as shown in Fig. 3. R_H appears to saturate above $H = 2$ kG at a value $R_H \simeq +1.7 \times 10^{-11}\ \Omega$ cm/G $= 1.7 \times 10^{-3}$ cm^3/C. The temperature dependence ($T < 100$ K) at 4.5 kG is shown in Fig. 4. R_H is constant above 20 K, remaining constant at 0.25×10^{-3} cm^3/C up to about 100 K at which point the signal is lost in the increasing noise. The Hall constant increases abruptly below 15 K to $+1.7 \times 10^{-3}$ cm^3/C below 5 K. The sign is positive indicating that R_H is dominated by positive charge carriers over the entire temperature range.

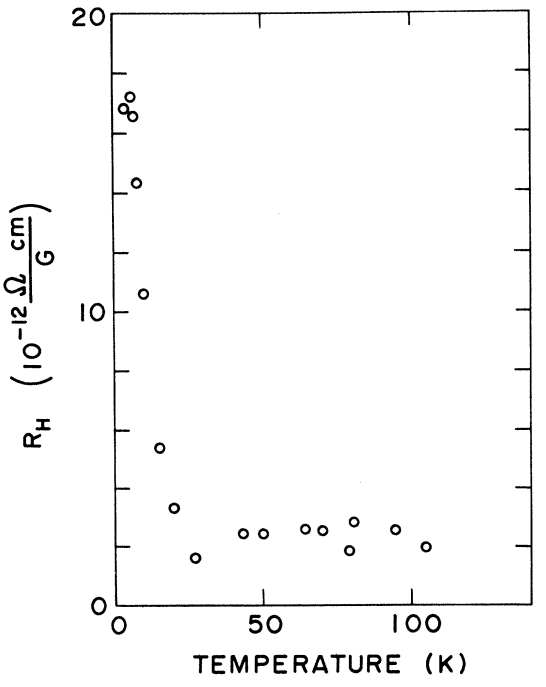

FIG. 4. Hall coefficient (R_H) vs temperature at 4.5 kG.

III. LOW-FIELD MAGNETORESISTANCE OF $Hg_{3-\delta}AsF_6$: THEORY

We consider a Fermi surface consisting of cylinders[6] with cross section described by planar sheets and circular corners of radius Δk (as sketched in Fig. 5), and calculate the magnetoconductivity tensor and the magnetoresistance. Within the planar part (Fig. 5) we assume

$$\epsilon = \frac{\hbar^3 k_i^2}{2m^*} \tag{1a}$$

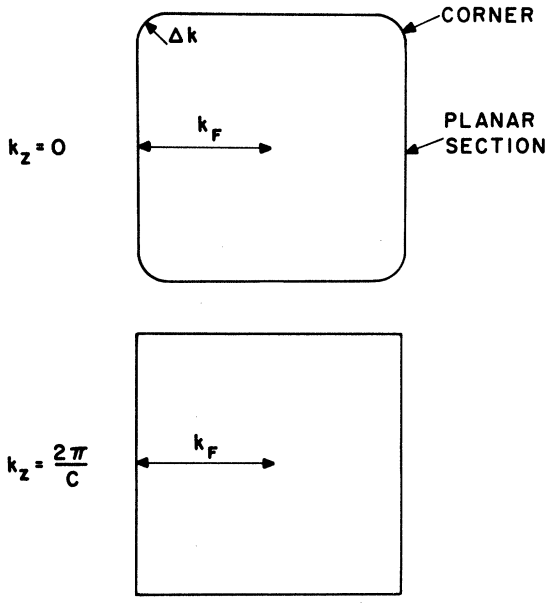

FIG. 5. Cross sections of the Fermi-surface cylinders at $k_z = 0$ and $k_z = 2\pi/c$ where c^* is the cylinder axis. The effect of the weak interchain coupling goes to zero at $k_z = 2\pi/c$ because of the symmetry of the band structure.

where $i = x,y$; whereas in the corner regions

$$\epsilon = \frac{\hbar^2\{[k_x-(1-\gamma)k_F]^2+[k_y-(1-\gamma)k_F]^2\}}{2m^*\gamma} \quad , \tag{1b}$$

where k_F is the one-dimensional Fermi wave vector defined by [Eq. (1)] $E_F = \hbar^2 k_F^2/2m^*$ where E_F is the Fermi energy. The effective mass, m^*, is assumed constant over the Fermi surface. The weak interchain coupling is described by the small parameter $\gamma \equiv \Delta k/k_F$, which describes the "sharpness" of the corner. We note explicitly that $\Delta k(k_z)$ is a function of k_z (z is the cylinder axis). From the symmetry of the crystal structure[5] we obtain an effective tight-binding matrix element[7] proportional to $\cos(k_z c'/2)$. Thus

$$\Delta k(k_z) = \Delta k^0 \cos\frac{k_z c'}{2} \quad \left(-\frac{\pi}{c'} < k_z < \frac{\pi}{c'}\right) \tag{2a}$$

or

$$\Delta k(k_z') = \Delta k^0 \sin\frac{k_z' c'}{2} \quad , \tag{2b}$$

where $k_z' c' = \pi - k_z c'$ and c' is the distance between parallel chains (along c); i.e., $c' = \frac{1}{2}c$ where c is the lattice constant. Note that $\Delta k(k_z)$ goes to zero at $k_z = 2\pi/c$ and takes on the maximum value Δk^0 at $k_z = 0$ (see Fig. 5).

In a magnetic field, the *local* cyclotron frequency can be defined as the angular rate of change of the group velocity

$$\omega_c(\vec{k}) \equiv \frac{d\varphi}{dt} \quad .$$

By this definition, $\omega_c(\vec{k}) = 0$ on the planar sections, whereas at the rounded corners, $\vec{k} = (\Delta k\cos\varphi, \Delta k\sin\varphi) + \vec{k}_0$,

$$\hbar\frac{d\vec{k}}{dt} = q\vec{v}(k)\times\vec{H} \quad , \quad \hbar\Delta k\frac{d\varphi}{dt} = \hbar\Delta k\,\omega_c(k) \quad ,$$

so that

$$\omega_c(k) = \frac{qH}{\hbar}\frac{v}{\Delta k} = \omega_c^0\left(\frac{k_F}{\Delta k}\right) \quad , \tag{3}$$

where $\omega_c^0 = qH/m^*$.

The dispersion relations, Eqs. (1a) and (1b), describe the two-dimensional limit of the three-dimensional model considered by Allgaier and Perl.[10] We assume in addition a two-relaxation-time model[2]; τ_p on the planar part and τ_c in the corner regions. The magnetoconductivity tensor can be written[11]

$$\sigma_{ij} = \sum_{n=0}^{\infty}\sigma_{ij}^{(n)}(H) \quad (i,j = x,y) \quad ,$$

$$\sigma_{ij}^{(n)}(H) = 2\frac{q^{(n+2)}}{(2\pi)^3\hbar^n}H^n\int v_i\left[\tau\left(v_x\frac{\partial}{\partial k_y} - v_y\frac{\partial}{\partial k_x}\right)\right]^n(v_j\tau)\left(-\frac{\partial f_0}{\partial\epsilon}\right)d^3k \quad , \tag{4}$$

where f^0 is the Fermi distribution function, q is the electron charge, $v_i(1/\hbar)\partial\epsilon/\partial k_i$, and the magnetic field is assumed to be along the z direction. Using Eq. (3), we find

$$\sigma_{xx}^{(0)} = \frac{q^2}{4\pi^3}\int v_x^2\tau\left(-\frac{\partial f_0}{\partial\epsilon}\right)d^3k = \frac{q^2}{4\pi^3}\int v_x^2\tau\frac{dl}{\hbar v_F}dk_z \quad ,$$

where v_F is the Fermi velocity and dl is an element of length along the Fermi surface in the xy plane. Substituting from the dispersion relations [Eq. (1)]

$$d\sigma_{xx}^{(0)} \simeq dn\frac{q^2\tau_p}{m^*} \quad , \tag{5}$$

where $dn \equiv (4k_F^2/4\pi^3)dk_z$ is the number of electrons in the differential cross section centered at k_z, and $m^* v_F \equiv \hbar k_F$. Integrating along k_z

$$\sigma_{xx}^{(0)} \simeq \frac{nq^2\tau_p}{m^*} \quad . \tag{6}$$

where n is the total number of electrons. Since the planar parts of the Fermi surface provide the dominant contribution to $\sigma_{xx}^{(0)}$, only τ_p occurs in Eqs. (5) and (6). The other components are calculated in a similar manner. Since all derivatives vanish on the planar sections,

$$\sigma_{xx}^{(2)} = \frac{q^4}{4\pi^3\hbar^2}H^2\tau_c^3\int_{\text{corner}}v_x^2\left(\frac{\partial v_y}{\partial k_y}\right)\left(-\frac{\partial v_x}{\partial k_x}\right)\frac{dl}{\hbar v_F}dk_z$$

$$= (-1)\frac{q^4}{4\pi^3\hbar^2}H^2\tau_c^3\int_{\text{corner}}v_x^2\left(\frac{\hbar}{m^*\gamma}\right)^2\frac{dl}{\hbar v_F}dk_z \quad .$$

Thus, using $v_x = v_F\cos\varphi$ and $dl = \gamma k_F d\varphi$

$$d\sigma_{xx}^{(2)} = -dn\frac{1}{\gamma}\frac{\pi}{4}\frac{q^2\tau_c}{m^*}(\omega_c^0\tau_c)^2 \quad , \tag{7}$$

where $d\sigma_{xx}^{(2)}$ is the contribution to $\sigma_{xx}^{(2)}$ from the differential cross section centered at k_z. Note that $d\sigma_{xx}^{(2)}$ is enhanced by the factor $\gamma^{-1} = k_F/\Delta k$. For σ_{xy}

$$\sigma_{xy}^{(1)} = \frac{q^3}{4\pi^3\hbar}H\int_{\text{corner}}v_x\tau_c^2 v_x\frac{\partial v_y}{\partial k_y}\frac{dl}{\hbar v_F}dk_z$$

and

$$d\sigma_{xy}^{(1)} = \frac{q^3}{4\pi^3\hbar} H \tau_c^2 \frac{v_F k_F}{m^*} \pi dk_z \ .$$

Again, $d\sigma_{xy}^{(1)}$ represents the contribution from the differential cross section centered at k_z. Thus

$$d\sigma_{xy}^{(1)} = dn \frac{\pi}{4} \frac{q^2 \tau_c}{m^*} (\omega_c^0 \tau_c) \ . \tag{8}$$

Note that for σ_{xy}, the enhancement factor is canceled by the smallness of the corner.

In a weak magnetic field, the magnetoresistance can be written in terms of the magnetoconductivity components

$$\frac{\Delta\rho_{xx}(H)}{\rho_{xx}(0)} = -\frac{\sigma_{xx}^{(2)}}{\sigma_{xx}^{(0)}} - \left(\frac{\sigma_{xy}^{(1)}}{\sigma_{xx}^{(0)}}\right)^2 \ . \tag{9}$$

Comparing Eqs. (7) and (8), for the Fermi surface shape of Fig. 5 ($\gamma << 1$)

$$\frac{\Delta\rho_{xx}(H)}{\rho_{xx}(0)} \simeq -\frac{\sigma_{xx}^{(2)}}{\sigma_{xx}^{0}} \ .$$

Thus, to complete the calculation

$$\frac{\Delta\rho(H)}{\rho(0)} = -\frac{\int d\sigma_{xx}^{(2)}}{\sigma_{xx}^{(0)}} \ .$$

When $\omega_c(k_z')\tau_c >> 1$, the magnetoresistance deviates from the weak-field behavior. From Eqs. (2) and (3), this always occurs near $k_z' = 0$. Experimentally, we find[2] that the magnetoresistance becomes approximately linear in H, when H exceeds the weak-field limit. For the small magnetic fields we are concerned with here ($H \sim 10$ Oe), both the number of electrons for which $\omega_c(k_z')\tau_c >> 1$ is very small, and the contribution to $d\sigma_{xx}^{(2)}$ has a weaker field dependence. We can therefore evaluate Eq. (10) by ignoring contributions from sections for the Fermi surface so close to $k_z' = 0$ that $\omega_c(k_z')\tau_c >> 1$;

$$\frac{\Delta\rho(H)}{\rho(0)} = -\frac{1}{\sigma_{xx}(0)} \int_{k_{z,\min}'}^{\pi/c'} d\sigma_{xx}^{(2)} \ ,$$

where $k_z'(\min)$ is the value for which $\omega_c(k_z')\tau_c = 1$; i.e.,

$$\Delta k(k_{z,\min}') = k_F \omega_c^0 \tau_c \tag{10}$$

or

$$k_{z,\min}' = \frac{2}{c'} \arcsin\left(\frac{k_F}{\Delta k^0} \omega_c^0 \tau_c\right) \simeq \frac{k_F}{\Delta k^0} \frac{(\omega_c^0 \tau_c)^2}{c'} \ .$$

The replacement of the arcsine by its argument is valid in the low-field limit. Thus, carrying out the integration

$$\frac{\Delta\rho(H)}{\rho(0)H^2} \simeq \left(\frac{\tau_c}{\tau_p}\right)\left(\frac{q\tau_c}{m^*}\right)^2 \frac{k_F}{2\Delta k^0} \ln\left[2\left(\frac{m^*}{qH\tau_c}\right)\frac{\Delta k^0}{k_F}\right] \ . \tag{11}$$

This expression holds only in the weak-field regime defined by $H < H_0$ where $H_0 = (\Delta k^0/k_F)(2m^*/q\tau_c)$.

At low magnetic fields the magnetoresistance is enhanced and varies as $H^2|\ln H|$. For comparison with experiment it is useful to rewrite Eq. (11) in the following form:

$$\frac{\Delta\rho(H)}{\rho(0)H^2} = \left(\frac{q\tau_p}{m^*}\right)^2 \frac{k_F}{2\Delta k^0} \left(\frac{\tau_c}{\tau_p}\right)^3 \ln\left[\left(\frac{2}{\omega_c^0\tau_c}\right)\frac{\Delta k^0}{k_F}\right] \ , \tag{12}$$

where τ_p is the scattering time appropriate to the flat planar sections which make up most of the Fermi surface and thus determine the conductivity in zero magnetic field.[2] Equation (12) implies an enhancement factor

$$\eta(H) \equiv \frac{\Delta\rho(H)}{\rho(0)H^2} \frac{1}{(\mu_T)^2} \ , \tag{13a}$$

$$\eta(H) = \frac{k_F}{2\Delta k^0} \left(\frac{\tau_c}{\tau_p}\right)^3 \ln\left[\left(\frac{2}{\omega_c^0\tau_c}\right)\frac{\Delta k^0}{k_F}\right] \ , \tag{13b}$$

where $\mu_T = (q\tau_p/m^*)$ is the transport mobility in the ab plane. The enhancement was noted earlier.[2] However the enhancement factor given in Eqs. (12) and (13a) differs from that obtained earlier as a result of the importance of including the dependence of Δk on k_z (and the explicit dependence on τ_c). This leads to the $\ln H$ dependence which is a principal point of this paper (see Fig. 2). The physics leading to the logarithmic dependence was not included in our earlier work, nor in that of Allgaier and Perl.[10] The extension of the theory to more than one band is straightforward. However, since the de Haas–van Alphen results[6] imply a series of cylinders along c with similar values for $\Delta k^0/k_F$ on each, the single band surface of Fig. 5 should provide an adequate starting point.

IV. DISCUSSION: THE TWO-RELAXATION-TIME MODEL AND THE INTERCHAIN COUPLING

The $\ln H$ dependence predicted by Eq. (12) is clearly evident in Fig. 2 for magnetic fields above about 2 G. Equations (12) and (13) imply

$$\frac{\Delta\rho(H)}{\rho(0)H^2} = A \ln\left(\frac{H_0}{H}\right) \ ,$$

where

$$A = \left(\frac{k_F}{2\Delta k^0}\right)\left(\frac{\tau_c}{\tau_p}\right)^3 \mu_T^2 \tag{14a}$$

and

$$H_0 = \frac{\Delta k^0}{k_F}\left(\frac{2m^*}{q\tau_c}\right) \ , \tag{14b}$$

$$AH_0 = \left(\frac{\tau_c}{\tau_p}\right)^2 \mu_T \ . \tag{14c}$$

From Fig. 2, one obtains $A \simeq 4 \times 10^{10}$ cm^4/V^2 s^2 and $H_0 \simeq 130$ Oe. Thus, from Eq. (14b) (with $\tau_c \sim 2 \times 10^{-12}$ s as determined below and $m^* = 0.35 m_e$) one finds $\Delta k^0/k_F \simeq 6 \times 10^{-3}$. Then, from Eq. (14c) (with transport mobility[2] $\mu_T \simeq 4.5 \times 10^4$ cm^2/V s), we obtain $(\tau_c/\tau_p) \sim 1$. The ratio of relaxation times is much closer to unity than the estimate inferred[2] earlier from the high-field magnetoresistance, suggesting that magnetic breakdown is important at high magnetic fields.

The logarithmic dependence down to 2 G implies, through Eq. (10), that $\Delta k(k_{z,\min})/k_F \approx 10^{-4}$; the corners of the Fermi surface are extremely sharp at this value of k_z implying the existence of a true cusp (in the physical sense). This is apparently the first time that a singularity of this kind has been observed in a Fermi surface. The existence of a low-field cutoff for the $\ln H$ dependence can be understood as a result of thermal smearing of the Fermi surface. When the field becomes so small that $\Delta k(k_z) < 2\pi/l$ where $l = v_F \tau_c$, the corner becomes smeared, and the $\ln H$ dependence is arrested. Thus, from Eq. (10), the saturation occurs when

$$\omega_c^0 \tau_c < \frac{\pi \hbar}{E_F \tau_c} \quad . \tag{15}$$

Using the experimental cutoff (~ 2 G) and taking $E_F = 3.5$ eV,[12] one finds $\tau_c \sim 2 \times 10^{-12}$ s. This value should be compared with the corresponding scattering time, τ_p, appropriate to the flat planar sections which make up most of the area of the Fermi surface, and thus determine the conductivity in zero magnetic field.[2,3] From the measured dc conductivity, making use of the number of carriers derived from the dHvA measurements[6] we obtained $\mu_T = 4.5 \times 10^4$ cm^2/V s for the mobility[2] at 4.2 K. Assuming an effective mass[6] $m^* \simeq 0.35 m_e$ implies that $\tau_p \simeq 10^{-11}$ s at 4.2 K. Thus $(\tau_p/\tau_c) \sim 1$, consistent with the results obtained from Eq. (14c).

The low-field data presented here, therefore, imply that a single relaxation-time model is adequate, and that $\tau_c \simeq \tau_p$ is uniformly long over the entire Fermi surface. Note that the relaxation time $\tau(k_i, k_F)$ is a function of both the initial and final values of k. Thus τ_p represents primarily backscattering processes, where an electron scatters from the $+k_F$ to the $-k_F$ *planar* sheet; τ_c included in addition perpendicular-chain scattering appropriate to values of $\vec{k}$ near a corner (Fig. 5). Evidently, such interchain scattering processes are weak.

The value obtained for the interchain coupling ($\Delta E/E_F \simeq 6 \times 10^{-3}$) is surprisingly small given that the point of closest approach of nearest-neighbor perpendicular chains is only 3.1 Å.[5] Thus, the low-field magnetoresistance implies extremely weak interchain coupling (even somewhat smaller than earlier estimates[2]) and provides an independent measure of the quasi-one-dimensionality of the electronic structure of $Hg_{3-\delta}AsF_6$.

Rice[13] has suggested that the long scattering time and correspondingly large mean free path in this quasi-1D system may result from many-body correlations due to electron-electron interactions such that $(g_1 - 2g_2) > 0$ where g_2 and g_1 are the forward and backward scattering amplitudes, respectively. In this case it has been shown that impurity backscattering goes to zero with decreasing temperature.[14] Alternatively, Weger[15] has noted that in the case of a long-range potential backscattering is small compared to processes that can carry an electron around the Fermi surface (by several small steps) in three dimensions. The bare potential, $V_0(k)$, due to a defect outside the chain may have a long range compared with $(2k_F)^{-1}$. Metallic screening would be expected to shorten the range and also reduce the magnitude, so that $V(k)$ may be weak for $k \sim 2k_F$. We note that a similar suppression of residual resistance has been reported recently for the quasi-one-dimensional conductor $(TMTSF)_2PF_6$.[16,17] More work is needed to settle this important question.

In spite of this very weak interchain coupling we expect that at sufficiently low temperatures (weak scattering) the wave functions will be coherent over many chains in the ordered 3D crystal structure, leading to a 3D band structure. Indeed, the results of Razavi *et al.*[6] confirm the existence of several electron and hole bands generated by Bragg scattering from the 3D crystal structure. From the energy-band structure deduced from the de Haas–van Alphen studies[6] we can calculate the hole and electron densities: $n_h = 3.0 \times 10^{21}$ cm^{-3} and $n_e = 0.5 \times 10^{21}$ cm^{-3}. At high enough magnetic

$$R_H = \frac{1}{(n_h - n_e) qc} \quad . \tag{16}$$

The result, $R_H(\text{theory}) = 2.5 \times 10^{-3}$ cm^3/C, is in reasonable agreement with the low-temperature experimental value, $R_H(\text{expt}) = 1.7 \times 10^{-3}$ cm^3/C. If we include also the δ and α bands observed by Razavi *et al.*,[6] the number of carriers is somewhat larger (by about 50%) giving a still closer agreement with the experimental value of R_H.

The field dependence of R_H implies saturation at about 2 kG. Saturation should occur when $\omega_c \tau > 1$ so that the carriers can sample the full Fermi surface. Since the full orbit is involved, $\tau^{-1} = \gamma \tau_c^{-1} + (1 - \gamma) \times \tau_p^{-1} \simeq \tau_p^{-1}$ where $\gamma = \Delta k/k_F$ is the fraction of the surface in the corner regions. Thus, from Fig. 3, $\omega_c^0 \tau_p > 1$ at 2 kG. Using the effective mass averaged over the Fermi surface as obtained by Razavi *et al.*,[6] $m^* = 0.35 m_e$, we find $\omega_c^0(2\text{ kG}) = 10^{11}$ and $\tau_p > 10^{-11}$ at 4.2 K, in agreement with the value estimated from the magnitude of the resistivity at 4.2 K ($\tau_p \sim 10^{-11}$ s).

The Hall coefficient decreases from $R_H = +1.7 \times 10^{-3}$ cm^3/C at the lowest temperatures to R_H

$= +0.25 \times 10^{-3}$ cm^3/C above 15 K. Since R_H at 4.2 K is rather strongly field dependent (Fig. 3), it is possible that we have a classical strong-field–to–weak-field transition; at strong field, Eq. (16) is valid, while in weak fields

$$R_H = \frac{n_h \mu_h^2 - n_e \mu_e^2}{qc(n_h u_h + n_e \mu_e)^2} \quad . \tag{17}$$

At 4.2, we can reach the strong-field value at a field of 2 kOe, since τ is long. Above 15 K, we cannot reach this limit since τ is much shorter. The high-T value is consistent with $\mu_e/\mu_h \simeq 2$, which is not unreasonable. Note that the positive carriers still dominate.

An alternative explanation to the large change in R_H, is a 3D to 1D crossover resulting from "localization" of wave functions onto 1D Hg chains. A similar increase was observed in HMTSF-TCNQ[18] near 40 K. As a result of the localization onto 1D chains, the wave functions change from 3D Bloch states, with quantum numbers (k_x,k_y,k_z) at low temperatures to 1D, with quantum numbers (x,k_y,z) at high temperatures, where y is the chain direction. The changeover occurs at $t_\perp \tau \simeq h$, where $t_\perp$ is the interchain transfer matrix element and τ is the appropriate electron-phonon relaxation time.

The situation for $Hg_{3-\delta}AsF_6$ is different because of the two perpendicular families of chains. In this case, the Bloch states (k_x,k_y,k_z) are defined as long as $\vec{k}$ can be localized to the corner region of size Δk. This requires that the wave function be coherent over $N = (a\,\Delta k)^{-1}$ parallel chains, where a is the lattice constant; an electron-phonon relaxation on any one of these chains will destroy the coherence and remove the electronic state from the corner region. Note that in the coherent-to-diffusive transition, there are two stages; first the smearing $\delta k > \Delta k$ where Δk is the size of the corner and second, $\Delta k > \pi/a$. For $\Delta k < \pi/a$, the electrons are not localized on individual chains, but on groups of N parallel chains where $N \sim (a\,\delta k)^{-1}$. To estimate the criterion for the first stage, we note that the probability that a carrier will not scatter in a time t it therefore given by $\exp(-Nt/\tau)$ so that the effective scattering time is $\tau/N = \tau(a\,\Delta k)$. Therefore in the case of interest here, the condition for a transition from a 3D Bloch state to localization onto 1D chains is given by

$$(\tau_p a\,\Delta k)\, t_\perp \simeq \hbar \quad , \tag{18a}$$

$$\tau_p (a k_F) \frac{t_\perp^2}{E_F} \simeq \hbar \quad . \tag{18b}$$

In Eq. (18) we have explicitly written $\tau = \tau_p$ where τ_p is the temperature-dependent scattering time associated with the individual mercury chains. Note that for crossed chains, a given value of $t_\perp$ has a much smaller effect in destroying the one-dimensional nature than for parallel chains. For crossed chains the relevant quantity is $t_\perp^2/E_F$, while for parallel chains, it is $t_\perp$. This feature also exists in the $A15$ superconductors.[19]

Above 20 K, in the region of 1D transport implied by the above analysis, the Hall coefficient remains positive. A straightforward analysis assuming four chains per unit cell with each AsF_6^- ion taking one electron leads to an electron density $n_e = 2.6 \times 10^{22}$ cm^{-3} or to a hole density $n_H = 4.7 \times 10^{21}$ cm^{-3}. Since the Hall effect is positive and the 1D Fermi surface (FS) is simply connected, we expect $R_H = +(n_h|q|c)^{-1} = 1.3 \times 10^{-3}$ cm^3/C whereas the measured value is $R_H = 0.25 \times 10^{-3}$ cm^3/C. Alternatively, using the experimental value implies a carrier density of 2.5×10^{22} cm^{-3} approximately equal to the electron density. It appears that although the sign is positive the magnitude of R_H is determined by the density of electrons in the 1D regime above 20 K.

In a purely 1D metal with chains in the x direction, $\sigma_{xy} = 0$, and $\sigma_{yy} = 0$. Therefore the Hall coefficient

$$\frac{\sigma_{xy}/H}{\sigma_{xx}\sigma_{yy} + \sigma_{xy}^2}$$

is not defined. However, for slightly nonplanar FS, neither σ_{xy} nor σ_{yy} vanish, and the classical value of the Hall coefficient is obtained (for a single relaxation time). This is the situation in the organic metals of the tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ) family. From experimental studies of parallel-chain materials, HMTSF-TCNQ[20] and TTF-TCNQ,[21] it has been demonstrated that the Hall constant has the classical value $R_H = (1/n_e c)(\mu_h - \mu_e)/(\mu_h + \mu_e)$ even for diffusive conductivity between chains. (In these systems, $n_e = n_h$, because the number of holes on the donor chain must equal the number of electrons on the acceptor chain to preserve charge neutrality.) For crossed chain systems, as in $Hg_{3-\delta}AsF_6$, σ_{xy} still vanishes when there is no coupling between the chains, but σ_{yy} does not. Therefore the Hall constant should vanish when there is no coupling between orthogonal chains. In the $Hg_{3-\delta}AsF_6$ case, there is some coupling. Unfortunately, there does not exist a detailed understanding regarding a system of crossed chains in the diffusive regime.

V. SUMMARY

As a result of the unusual Fermi surface associated with the two sets of perpendicular chains in $Hg_{3-\delta}AsF_6$, the low-field magnetoresistance is enhanced and a $\ln H$ dependence for $\Delta\rho(H)/\rho(0)H^2$ is predicted. The $\ln H$ dependence has been observed in high sensitivity low-field experiments described in this paper. Analysis of the results indicates that a single relaxation-time model is adequate and provides

an estimate of the interchain coupling $\Delta E/E_F \simeq 6 \times 10^{-3}$. The value for τ_p/τ_c is of order unity; considerably smaller than the estimate obtained from the high-field magnetoresistance[3] suggesting that magnetic breakdown is important at high fields. The sign and magnitude of the low-temperature Hall effect are consistent with the 3D band structure inferred from de Haas–van Alphen measurements. However, above 20 K, the results are anomalous; the sign of R_H is positive, but the magnitude appears to result from the density of electrons in the 1D band structure.

The low-field galvanomagnetic studies, therefore, lead to a picture of weakly coupled chains ($\Delta E/E_F < 10^{-2}$) qualitatively consistent with the linear chain structure of $Hg_{3-\delta}AsF_6$. However, the extremely small coupling is surprising given that the point of closest approach of nearest-neighbor perpendicular chains is only 3.1 Å, since at this distance the transfer integral between two Hg atoms is estimated to be greater than 1 eV.[22] A more detailed theory of the band structure, including the effects of incommensurability of the wave functions,[23] may be required.

The absence of residual resistivity remains an important aspect of the problem. The suggestion that this might be the result of many-body correlations in one dimension ($g_1 - 2g_2 > 0$) with the implication of triplet superconductivity at lower temperatures is of particular interest.

Note added in proof. Our value of $\Delta E/E_F$ depends on the measured value of ω_c in high magnetic fields ($H \simeq 4.5$ T), where magnetic breakdown may lead to some large orbits. If the cross-sectional areas of the Fermi surface in small fields are smaller, ω_c in small fields will be larger, and $\Delta E/E_F$ will as a result be larger too.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation—Materials Research Laboratory Program (DMR-7923647). We are grateful to Dr. Robert Allgaier for reading the manuscript carefully and critically and for his constructive suggestions. We thank F. Razevi and F. Mueller for stimulating discussions. Partial support for M. Weger was obtained from the Penn-Israel Exchange Fund.

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