Hall effect in the two-dimensional metal Sr_2RuO_4

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We report a detailed study of the Hall effect in Sr_2RuO_4, the first layered perovskite superconductor which does not contain copper ($T_c \cong 1$ K). The Hall coefficient (R_H) was measured at temperatures between 20 mK and 300 K, and an unusual dependence of R_H on the applied magnetic field was observed. R_H has a strong temperature dependence below 25 K, but below 1 K it saturates at a value of -1.15×10^{-10} m^3/C. The Fermi surface of Sr_2RuO_4 is known from quantum oscillation measurements, and since it is nearly two dimensional, it is possible to derive a simple expression for R_H using methods developed by Ong [Phys. Rev. B **43**, 193 (1991)]. We show that if the mean free path, l, is assumed to be isotropic at low temperatures, it is possible to make an accurate quantitative calculation of R_H on the basis of the known Fermi surface parameters. [S0163-1829(96)08131-3]

I. INTRODUCTION

Metallic behavior was reported in Sr_2RuO_4 many years ago,[1] and detailed measurements of the resistivity (ρ) were performed on single crystals in the years following the discovery of high-temperature superconductivity in the cuprates,[2] motivated by the fact that it has essentially the same structure as $La_{2-x}(Ba,Sr)_xCuO_4$, and might itself be a candidate for superconductivity. Initially, none was found, and interest in Sr_2RuO_4 was mainly due to its potential as a highly conducting component in layered devices based on the cuprates. Recently, however, Maeno and coworkers showed that it is indeed a superconductor, with a low transition temperature of approximately 1 K.[3] Subsequent investigations have established that unlike the cuprates, Sr_2RuO_4 has low-temperature properties that are consistent with the predictions of Fermi-liquid theory, although it is extremely anisotropic, with $\rho_c/\rho_{ab}>500$ at low temperatures.[4,5] Both band-structure calculations[6,7] and direct Fermi-surface measurements via the de Haas–van Alphen and Shubnikhov–de Haas effects[5] show that the Fermi surface consists of three essentially cylindrical sheets, two of which are electronlike and one of which is holelike.

Very soon after the discovery of the superconductivity, Shirakawa *et al.*[8] showed that the Hall coefficient (R_H) of Sr_2RuO_4 has a complicated temperature dependence, as might be expected from a material containing electronlike and holelike carriers, and that the sign of R_H at temperatures below approximately 30 K is negative. In this paper we report the results of a detailed study of the Hall effect between 20 mK and 300 K, and show that the Hall voltage, V_H, is nonlinear in field at low temperatures. The normal-state value of R_H can be obtained below the superconducting transition temperature (T_c) by working at fields larger than the upper critical field (H_{c2}), which is only 0.04 T when the field is applied along the c direction. We confirm the very strong temperature dependence between 2 and 50 K first reported by Shirakawa *et al.*, but observe that below 1 K, R_H becomes almost temperature independent as we enter a regime in which elastic impurity scattering is the dominant scattering mechanism. Using knowledge of the size of the Fermi-surface sheets from the quantum oscillation measurements,[5] and the assumption that the mean free path, l, is isotropic in the elastic-scattering limit, we can make an accurate quantitative prediction of the measured low-temperature value of R_H, and account qualitatively for its field dependence. We also suggest that the strong temperature dependence of R_H at higher temperatures could be a consequence of rather small temperature dependent changes in the **k** dependence of l.

II. EXPERIMENT

The sample growth is described in Refs. 3–5. For these measurements we used a single crystal (approximate dimensions 1 mm×0.5 mm×30 μm) whose high quality is evidenced by the fact that clear Shubnikhov–de Haas oscillations were observed in its magnetoresistivity. Low-temperature measurements between 20 mK and 6 K were performed in a dilution refrigerator/18 T magnet system by sweeping the magnetic field at constant temperature with a fixed sample position, and then repeating for the opposite field polarity, as described in Ref. 9. To operate above 1.2 K, most of the $^3He/^4He$ mixture was removed, with the remainder providing a thermal link with the 1.2 K pot, and a heater was used to reach temperatures up to 6 K. The sample was also studied in a conventional ^4He crysostat, at temperatures between 2.5 and 300 K, by rotating in a fixed field of 2 T

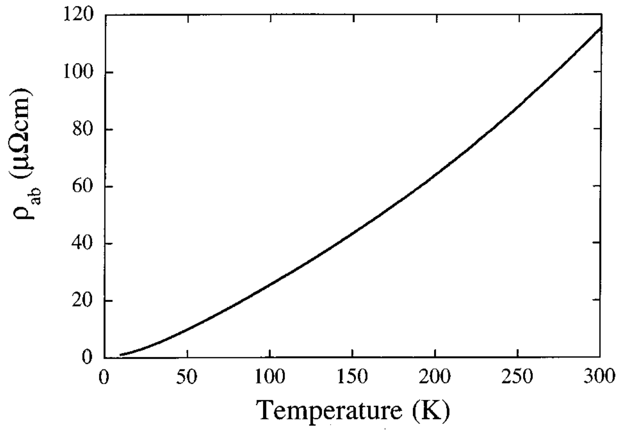

FIG. 1. The in-plane resistivity ρ_{ab} of the high-quality single crystal of Sr_2RuO_4 used in this experiment. The resistivity ratio $[\rho_{ab}(300\ K)/\rho_{ab}(4.2\ K)]$ is in excess of 100, and the sample showed Shubnikhov–de Haas oscillations (Ref. 5).

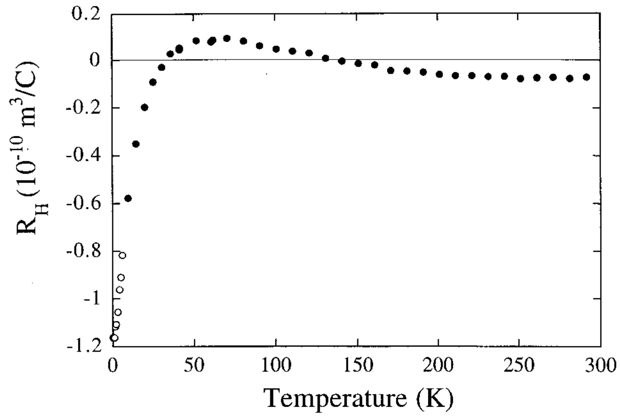

FIG. 2. The temperature dependence of the weak-field Hall coefficient R_H. The closed symbols show data taken by rotating the sample in a fixed field of 2 T in a ^{4}He cryostat, and the open ones show data obtained by reversing the magnetic field in a cryostat equipped with a dilution refrigerator (see text).

(see, for example, Ref. 10 for more details). The field dependence of V_H was checked at all temperatures in the 18 T system, and at 10, 45, 100, and 300 K in the ^{4}He cryostat. In all cases the magnetic field was applied parallel to the c axis of the crystal.

III. RESULTS AND DISCUSSION

A. Resistivity

The zero-field ab plane resistivity is shown in Fig. 1 at temperatures between 10 and 300 K. Below about 25 K, both ρ_{ab} and ρ_c vary as T^2 to high accuracy;[4] above this temperature, ρ_{ab} rises monotonically, with curvature which is much less strong than T^2. In this range, it can be modelled well using a weighted sum of T and T^2 contributions, with a strong T component, reminiscent of the behavior seen in the overdoped cuprates such as $La_{2-x}Sr_xCuO_4$ (Ref. 11) and $Tl_2Ba_2CuO_{6+\delta}$.[9,12] Although ρ_c shows a pronounced maximum at about 100 K,[3] no equivalent feature is seen in ρ_{ab}.[2,13] The sample studied here has a residual resistivity of approximately 0.7 $\mu\Omega$ cm and a resistivity ratio $\rho_{ab}(300\ K)/\rho_{ab}(4\ K)$ in excess of 100. It has often been noted that the extrapolated resistivity ratio of the stoichiometric cuprate $YBa_2Cu_3O_7$ is very high, but since this involves extrapolation over a wide temperature range of approximately 100 K, there has been some uncertainty. Some authors have argued that the very high ratio that is inferred is surely not plausible in a complex oxide material, and that it must be taken as evidence for solitonic excitations in the copper-oxygen planes which are not subject to the usual scattering processes.[14] In Sr_2RuO_4, it seems clear that the ground state is a Fermi liquid, so the observation of such a low residual resistivity is a direct demonstration of the kind of material quality that is possible in complex oxides in which the conductivity is not induced by the introduction of random stoichiometric defects.

B. Hall coefficient: Temperature and field dependence

In Fig. 2 we show the temperature dependence of R_H measured between 20 mK and 300 K with a fixed field of 2 T applied parallel to the c axis. These findings are similar to those reported in Ref. 8, although we are able to resolve a return to a negative value of R_H at temperatures above 130 K. The measurements were made in the relatively low field of 2 T because, as shown in Fig. 3, the Hall voltage, V_H, has an unusual *superlinear* field dependence at low temperatures (Fig. 3), which can be modeled well as a sum of linear and cubic terms. As can be seen in Fig. 3, the correction due to the higher power term is very small at fields up to 2 T. Field sweeps performed at more than ten temperatures below 6 K, and at 10, 44.5, 116, and 300 K confirmed that the superlinear enhancement is reduced as the temperature increases, so data taken at 2 T give a good estimate of the weak-field Hall coefficient over the whole temperature range studied. The origin of the unusual field dependence will be discussed in Sec. III D below.

C. Low-temperature Hall coefficient in the isotropic-l approximation

Details of the low-temperature behavior of R_H are presented in Fig. 4. Although the temperature dependence be-

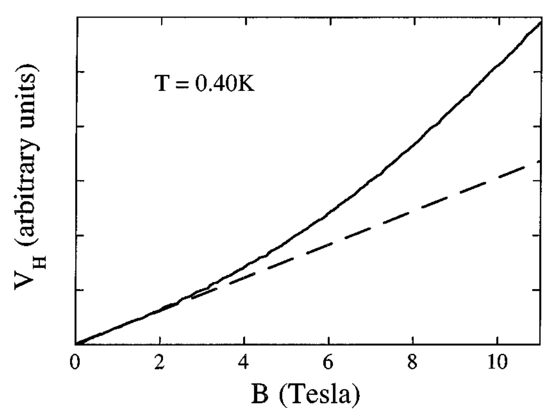

FIG. 3. Detailed measurements show that the Hall voltage, V_H, is nonlinear in field. The dashed line shows the gradient of the linear region at low field which can be used to deduce R_H.

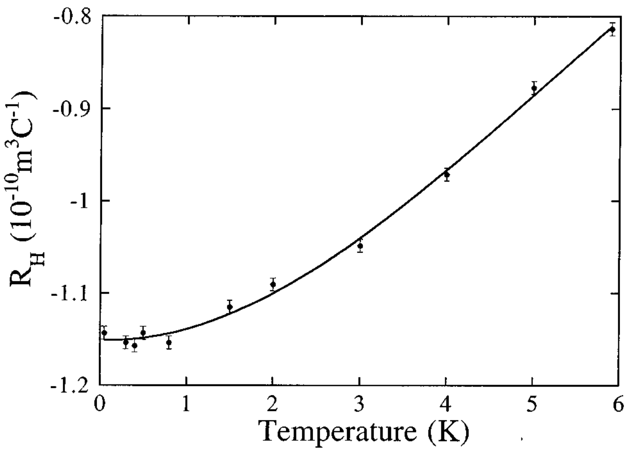

FIG. 4. The low-temperature behavior of R_H. Note that in spite of the strong temperature dependence between 1.5 and 30 K, R_H becomes essentially temperature independent below 1 K, indicating entry into the regime dominated by scattering from static impurities. The error bars were estimated from the error in fitting a line to the data below 2 T in field sweeps such as that shown in Fig. 3. The line is a guide to the eye.

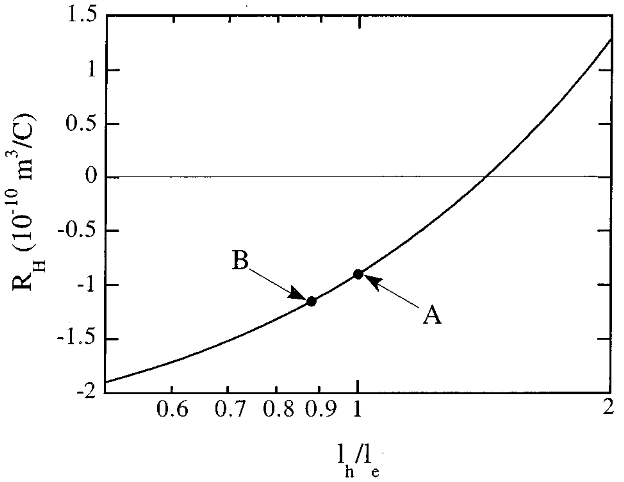

FIG. 5. The dependence of R_H as calculated using expression (1) on the ratio of the mean free path for the holes (l_h) to that for the electrons (l_e) which is assumed for simplicity to be equal for the two electron pockets.

tween about 1.5 and 50 K is very strong, R_H becomes essentially temperature independent below 1 K. This gives good evidence that we are reaching the regime in which large-angle elastic scattering from impurities dominates the transport properties. In this regime, it is common to assume that, to a first approximation, the mean free path l depends only on the separation between the impurities which provide the scattering centers, and is no longer sensitive to differences in the Fermi velocity either at different parts of the same Fermi surface sheet or on different sheets in the case of a multiband metal. This is known as the ''isotropic- l'' approximation. The value of l deduced from the resistivity in this approximation is ~1500 Å below 1 K, a figure which is consistent with estimates that can be made from the field dependence of quantum oscillations in other crystals from the same batch.[15]

In order to understand the low-temperature value of R_H, we use the elegant geometrical interpretation of the weak-field Hall effect in two dimensional metals in terms of the wave-vector dependence of $l=\mathbf{v}_F\tau$ derived by Ong.[16] Taking $R_H=\sigma_{xy}/B\sigma_{xx}^2$, where B is the magnetic field,[17] and assuming that the three pockets are almost circular with a **k** independent l in any individual pocket, we obtain a general expression for R_H in a multiband material:

$$R_H=\frac{2\pi d\Sigma_i(-1)^{n_i}l_i^2}{e(\Sigma_i k_F^i l_i)^2}, \tag{1}$$

where d is the interplane separation, and $n_i=1$ if Fermi surface sheet i is electronlike and 2 if it is holelike.[18] If we make the extra assumption that at sufficiently low temperature l is the same for all the pockets, the expression simplifies to

$$R_H=\frac{2\pi d\Sigma_i(-1)^{n_i}}{e(\Sigma_i k_F^i)^2}. \tag{2}$$

Using the known values of k_F of 0.3, 0.62, and 0.75 Å^{-1} for the hole pocket and the two electron pockets, respectively,[5] we obtain a prediction within the isotropic-l approximation of $R_H=-0.9\times10^{-10}$ m^3/C. This compares well with the measured value of -1.15×10^{-10} m^3/C, and indicates that the isotropic-l approximation is a reasonably good one as $T\rightarrow0$. To our knowledge, the combination of complete low-temperature Fermi surface and Hall effect data in a layered metal is unique, so this is the first time that such a direct test of the approximation has been possible.

That the agreement between the calculated and measured values is impressive can be better appreciated by considering Fig. 5, which gives the dependence of R_H calculated from expression (1) on the assumption (purely for illustration) that l is the same for the two electron pockets but different for the much smaller hole pocket. In this very simple model, the calculated value of R_H is extremely sensitive to the ratio l_h/l_e. Point A represents the calculated value if l is isotropic, and point B represents the measured value, which could be produced, for example, by l_h being only 10% smaller than l_e. If the effect of finite temperature were to reduce l_e more rapidly than l_h, the zero of R_H seen at about 30 K could be the result of l_h/l_e becoming only as large as 1.4. Although this simple model is only one example, it serves to illustrate that even in a material which is only partially compensated (i.e., $n_e\neq n_h$), the value of R_H can be extremely sensitive to details of the **k**-dependent scattering, and strong temperature dependences are likely.

D. Low-temperature analysis of the field dependence of the Hall coefficient and voltage

It is also possible to analyze the field dependence of V_H (Fig. 3) within the isotropic-l approximation. As the field is raised, and we leave the strictly weak-field regime, we have to take into account the higher-order terms in the Zener-Jones expansion. Taking the next higher-order term into account, the Hall voltage is given by $V_H=R_HB=(\sigma_{xy}+\sigma_3)/\sigma_{xx}^2$, where σ_3/σ_{xx}^2 is the next term in the expansion (proportional to B^3), given for a single band in the isotropic-l approximation by

$$\frac{\sigma_3}{\sigma_{xx}^2}=\frac{2\pi d(\omega_c\tau)^2(-1)^n}{ek_F^2}, \tag{3}$$

where ω_c is the cyclotron frequency and $n=1$ for an electron pocket and 2 for a hole pocket. Thus for a single band, σ_3 always has the opposite sign to σ_{xy}. Writing $\omega_c=ev_F\tau B/\hbar k_F$, we can extend to the multiband case:

$$\frac{\sigma_3}{B\sigma_{xx}^2}=\frac{-2\pi d(eBl)^2[\Sigma_i(-1)^{n_i}(1/k_F^i)^2]}{e\hbar^2(\Sigma_i k_F^i)^2}. \tag{4}$$

Now, the contribution from each band is seen to be weighted by the factor of $(1/k_F^i)^2$, favoring the contribution of small pockets. Taking our values of k_F^i for Sr_2RuO_4, we find that the contribution from the smaller hole pocket dominates the B^3 term and reverses the sign of σ_3/σ_{xx}^2, thus explaining the superlinear behavior seen in V_H. A fit containing only terms linear and cubic in B fits the field dependence well, and using $l\sim1500$ Å, we calculate the higher-order term to be substantial (0.4 times the linear term at 11 T, instead of the observed value of approximately 0.8). This agreement is good for a higher-order term—the factor of 2 discrepancy between the calculated and observed values could be the result of only 15–20 % deviations from isotropic-l, so the approximation is again seen to be a fairly good one at sufficiently low temperatures.

E. Difficulties of applying a simple analysis to high-temperature behavior

It should also be possible to interpret R_H at high temperatures, where the scattering lifetime τ rather than the mean free path l is isotropic.[16] As can be seen from Fig. 2, R_H is almost temperature independent above 200 K, and it is tempting to extend the analysis to that temperature range. However, if one naively attempts to estimate the ratio of mean free paths in Eq. 1 by taking $l_i=v_F^i\tau$, $v_F^i=\hbar k_F^i/m_i^*$, and using the low-temperature cyclotron masses of $3.2m_e$, $6.6m_e$, and $12m_e$ for the three pockets,[5] one obtains a value of approximately -0.5×10^{-10} m^3/C, in fairly poor agreement with the experimental value of -0.1×10^{-10} m^3/C. There could be several reasons for this. First, even if the band structure has the same meaning at these elevated temperatures, it is by no means clear that the low-temperature mass enhancements that were used in the estimation of v_F^i will survive to high temperature. However, as reported in Ref. 4, the Pauli susceptibility remains essentially temperature independent up to 700 K. A more likely explanation is that the method used for deducing v_F^i can only give a value averaged over each pocket. If there is a significant **k** dependence and the pockets are not perfectly circular, this is likely to affect the calculated value of R_H far more in the isotropic-τ regime than in the isotropic-l regime.

F. Magnetic contributions to the Hall effect?

It is well known that paramagnetic and ferromagnetic materials can have a large magnetic contribution to the Hall effect due to processes such as skew scattering or side jump. Normally, these effects are strong in materials whose paramagnetism results from local moments which give a strong Curie term in the susceptibility χ, and the magnetic contribution to the Hall effect has a very similar temperature dependence to that of χ.[19,20] Experiments on heavy-fermion materials show that if the paramagnetism is not Curie-like, the situation is different. In many of these compounds, the onset of coherence at low temperatures is accompanied by a dramatic reduction in the magnitude of the magnetic contribution to R_H, even though the large value of χ does not change very much in this temperature range.[20] In Sr_2RuO_4, χ is approximately 10^{-3} emu/mol, but it is essentially temperature independent, with little or no Curie term. Also, the field dependence that we see is superlinear all the way up to 18 T, and can be modeled using terms derived purely on the basis of the orbital Hall effect in a high-purity metal. We see no sign of the saturation that is commonly observed when there is a large magnetic contribution to the Hall effect. For these reasons, we believe that the data presented in Fig. 2 are dominated by the standard orbital R_H, although we cannot rule out some magnetic contribution.

IV. CONCLUSIONS

In conclusion, we have studied the Hall effect in Sr_2RuO_4 over a wide temperature range. The simplicity of its Fermi surface means that it is an ideal material in which to test the theoretical relationship between Fermi surface topology and the weak field Hall effect, and we have shown that it is possible to make a good estimate of the magnitude of the low-temperature Hall coefficient within the isotropic-l approximation.

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