Two-carrier Magnetoresistance: Applications to Ca$_3$Ru$_2$O$_7$

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Ambipolar transport is a commonly occurring theme in semimetals and semiconductors. Here we present an analytical formulation of the conductivity for a two-band system. Electron and hole carrier densities and their respective conductivities are mapped into a two-dimensional unit-less phase space. Provided that one of the carrier densities is known, the dimensionless phase space can be probed through magnetoresistance measurements. This formulation of the two-band model for conductivity is applied to magnetoresistance experiments on Ca$_3$Ru$_2$O$_7$. While previous such measurements focused on the low-temperature limit, we cover a broad temperature range and find negative magnetoresistance in an intermediate interval below the electronic transition at 48 K. The low-temperature magnetoresistance in Ca$_3$Ru$_2$O$_7$ is consistent with a two-band structure. However, the model fails to describe the full temperature and magnetic field dependence. Negative magnetoresistance found in an intermediate temperature range is, for example, not captured by this model. We thus conclude that the electronic and magnetic structure in this intermediate temperature range render the system beyond the most simple two-band model.

1. Introduction

The combination of electron and hole charge carriers is a common theme of condensed matter physics. It is also essential for many applications such as semiconductor junctions, excitonic solar cells and light emitting diodes. Transport properties of semimetals are defined by electron and hole charge carriers. In its simplest form, a semimetal consists of just one electron- and one hole-like band crossing the Fermi level. Exposed to a magnetic field, the electrons and holes experience a Lorentz force which results in a negative or positive Hall effect depending on whether conductivity is dominated by electrons or holes. Compensated ambipolar transport properties stem from the detailed balance between electron and hole mobilities. Most semimetals/metals have more than two bands crossing the Fermi level. Nevertheless, a two-band model can still be of some relevance and is widely used to discuss transport experiments. In some multiband systems, only two Fermi surface sheets host high-mobility carriers – making the two-band model approximately viable. Orbital selectivity found in systems with strong Hund's coupling may also trigger selective Fermi surface gapping while other sheets are untouched – thus providing another route to two-band physics. Finally, electronic reconstruction of single band systems is a common motif that can generate two-band structures. The wide applicability justifies a detailed analysis of the two-band model.

Here we show that the electron and hole transport properties can be described by two dimensionless parameters $\alpha$ and $\beta$. This ($\alpha, \beta$) phase space connects smoothly the ambipolar transport with the mono-carrier dominated regime. For constant filling, the system must move along ($\alpha, \beta$) specific contour lines. If the carrier density of - say the electrons – is known from angle resolved photoemission spectroscopy (ARPES) or quantum oscillation experiments, the temperature dependent parameters $\alpha$ and $\beta$ can be determined from a magnetotransport experiment. We apply this new formulation of the two-band model to magnetoresistance experiments of the semimetallic phase of Ca$_3$Ru$_2$O$_7$. While previous studies focused entirely on the low-temperature limit, we cover a broad temperature range and find negative magnetoresistance for 30 $K < T < 48$ K. For selected temperatures ($T \approx 10$ K), the observed low-field magnetoresistance is captured well by the two-carrier model. However, as a function of temperature (warming) the ($\alpha, \beta$) parametrisation is inconsistent with a constant filling. For the case of Ca$_3$Ru$_2$O$_7$, the two-band model fails to explain the full temperature and magnetic field dependence of the magnetoresistance. The most pronounced limitation of the two-band model is its inability to explain the negative magnetoresistance found for 30 $K < T < 48$ K. We discuss these discrepancies in terms of a two-stage Fermi surface reconstruction. Below 48 K, the system is a semimetal but with a Fermi surface including more than two sheets. A second reconstruction across 30 K simplifies the Fermi surface such that it hosts only a single electron- and hole-like pocket. Within this structure reasonable consistency with the two band model is reached at low temperatures.

2. Methods

High quality single crystals of Ca$_3$Ru$_2$O$_7$ were grown by the floating zone techniques. The orthorhombic ($Bb_{2}m$) crystals were detwinned, by pressing along an orthorhombic axis, using a thermo-mechanical detwinning device in combination with a polarized light microscope. The longitudinal resistivity (along the $b$-axis), Hall, magnetization and thermoelectric measurements were carried out in a Quantum Design Magnetic Property Measurement system and a Physical Property Measurement System with magnetic field $H$ applied along the crystallographic $c$-axis. For the thermoelectric experiments, temperature gradients along the orthorhombic $b$-axis (the longer lattice parameter) were recorded using Cernox chips.
with previous studies, the low-temperature positive magnetoresistance is consistent with isotherms (sharp sign change of the Seebeck coefficient associated with a Fermi surface reconstruction, signaled by a temperature scale) for temperatures as indicated – are also consistent with previous experiments. Hall resistivity $\rho_{xx}$ versus magnetic field for temperatures as indicated. The inset shows the temperature evolution of the Hall coefficient, $R_H$. (d) Magnetoresistance plotted as $[\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0)$ versus magnetic field squared for temperatures as indicated. Solid grey lines are linear fits.

3. Results

In Figs. 1a,b, we are plotting the zero magnetic field resistivity $\rho$ (along $b$ - axis) and thermopower $S$ versus temperature. The temperature dependence of both curves are in agreement with all previous experiments.\textsuperscript{23-25} Resistivity displays a weak drop across the AFM-a Néel ordering temperature $T_N = 56$ K and a sharp upturn below the spin-flip transition $T_s = 48$ K into the AFM-b phase. The latter transition is associated with a Fermi surface reconstruction, signaled by a sharp sign change of the Seebeck coefficient\textsuperscript{24} (Fig. 1b).

Below 30 K, the resistivity recovers a metallic Fermi-liquid-like ($\rho \propto T^2$) temperature dependence (Fig. 1a). Hall resistivity isotherms ($\rho_{xy}$ versus magnetic field $H$) – shown in Fig. 1c for temperatures as indicated – are also consistent with previous reports. Negative Hall resistivity is found for $T < T_s$.\textsuperscript{26}

Magnetoresistance plotted as $[\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0)$ versus $H^2$ is displayed in Fig. 1d for temperatures as indicated. The low-temperature positive magnetoresistance is consistent with previous studies,\textsuperscript{24,27} all of which showed, $[\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0) = (\mu_{MR} H^2)$ with mobility $\mu_{MR} \approx 0.1$ T$^{-1}$. Here we focus on the temperature dependent magnetoresistance in Ca$_3$Ru$_2$O$_7$. For $T > T_s$, a small positive magnetoresistance is found whereas for 30 K < $T < T_s$ it is negative before turning to large positive values in the $T \to 0$ limit. The derivative $d\rho_{xx}/dH$ is shown in Fig. 2a as a function of temperature and magnetic field. The onset of negative magnetoresistance and history dependent magnetic susceptibility (Fig. 2b) clearly correlate. Both effects appear below $T_s$.

The low-temperature magnetization and Hall resistivity isotherms display different field dependence (Fig. 2c). The magnetization $M$ is linear in magnetic field whereas $\rho_{xx}$ is displaying non-linear field dependence suggesting that these two quantities are not directly coupled. The linear trend of the low temperature magnetization data (Fig. 2a) excludes the possibility of an anomalous Hall effect. We also point out that although $\rho_{xx} \propto H^2$ in the low-field ($H \to 0$) regime, significant deviations are observed in larger magnetic field (see Fig. 2d and Reference.\textsuperscript{27}). In what follows, we discuss the low-temperature large positive and intermediate-temperature negative magnetoresistance behaviour within the two-band model.

4. Modelling

For metals with two Fermi surface sheets of different band curvature, the magnetoresistance is given by:$$\rho_{xx}(H) = \frac{\sigma_h^2 R_h + \sigma_e^2 R_e + \sigma_h \sigma_e R_h R_e}{R_h + R_e} H^2.$$(1)

The model consists of four parameters $\sigma_e$, $\sigma_h$, $R_e$, and $R_h$. The hole and electron conductivities are denoted by $(\sigma_h, \sigma_e)$ and the hole and electron Hall coefficients are indicated by $(R_h, R_e)$. Given that $\sigma_i = n_i e \mu_i$ and $R_i = \pm 1/(n_i e)$ where $i = e, h$, the model can be also expressed in terms of electron and hole carrier density ($n_e, n_h$) and mobility ($\mu_e, \mu_h$). This model generally assumes that the mobilities are independent of the applied magnetic field. Analysis of $\rho_{xx}$ and $\rho_{xy}$ isotherms in terms of the four parameters $(\sigma_e, \sigma_h, R_e, R_h)$ is typically associated with ambiguity.\textsuperscript{6} Although $\sigma_e$ and $\sigma_h$ are coupled parameters, since in zero-field, $\rho_{xx} = (\sigma_e + \sigma_h)^{-1} = \sigma^{-1}$, the
problem has three free parameters.

To make progress, we assume that the electron carrier density \( n_e \) is known, for example, from ARPES\(^{16}\) or quantum oscillation\(^{17}\) experiments. The problem then reduces to two equations with two unknowns (\( \sigma_h \) and \( n_h \)), lifting the ambiguity. In the low-field \( H \to 0 \) limit, the problem can be expressed in terms of two dimensionless parameters \( \alpha \) and \( \beta \):

\[
\alpha = \frac{R_H}{|R_e|} = \frac{\rho_{xy}}{H|R_e|} = \frac{\sigma_h^2 R_h/|R_e| - \sigma_e^2}{\sigma^2},
\]

and

\[
\beta = \frac{3C}{\sigma^4|R_e|^2} = \frac{3\sigma_h \rho_e}{(\sigma_h^2 R_h/|R_e| + \sigma_e^2)}.\]

with \( C \) defined by \( \rho_{xx} = \rho_0 + CH^2 \) where \( \rho_0 \) is the residual resistivity. Solving with respect to \( \sigma_h \) and \( R_h \) yields \( \sigma_h/\sigma = \mathcal{F}(\alpha, \beta) \), \( R_h/R_e = \mathcal{G}(\alpha, \beta) = [\alpha + (1 - \mathcal{F})^2]/\mathcal{F}^2 \) and \( \mu_h/\mu_e = \mathcal{G}/\mathcal{F} \), while \( \mathcal{F} \) and \( \mathcal{G} \) are all measurable quantities, this provides a test as to whether a two-band model is applicable. We notice that this is independent of our initial input for \( R_e \). Second, constant filling implies that \( n_e + n_h = n_e (1 + G^{-1}) \) is invariant. Therefore, if \( n_e \) remains constant, the system must stay on a \( G \)-contour. These contour lines "flow" from the electron to the hole dominated limit via the ambipolar\(^{2}\) regime (see Fig. 3a).

For example, increasingly negative Hall coefficient implies \( \alpha \to -1 \) and hence \( \beta \) should decrease for the system to stay on a constant \( n_e/n_h \) contour. Since both \( \sqrt{\beta} \) and \( \sigma \) scale with \( 1/R_e \), the shape of the contour lines are independent of \( R_e \). A third solution property stems from the \( n_e/n_h = 1 \) contour line\(^{29-31}\) given by \( \beta = 3(1 - \alpha^2)/4 \). If a system is known to be hole-doped, i.e. \( n_h > n_e (G < 1) \), it must satisfy \( |\alpha| < \sqrt{3}/2 \) and \( \beta < 3(1 - \alpha^2)/4 \). Deviation from any of the three properties implies: (a) non-constant filling, (b) the system is not having a two-band structure or (c) the two-band model assumptions are too simplistic.

5. Discussion

Before analysing the magnetoresistance of Ca\(_3\)Ru\(_2\)O\(_7\), we start by illustrating the use of the aforementioned solution properties. In doing so, we consider results obtained on hole and electron doped cuprates. For hole doped YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\), and YBa\(_2\)Cu\(_4\)O\(_8\), quantum oscillation experiments yields an
electron-like Fermi surface sheet with an area corresponding to \( R_c = -29\ \text{mm}^2 \). Since the system is hole-doped, we expect \( n_h > n_e \). Hall effect experiments suggest that \( \alpha \approx 1\).\(^{27}\) Already here a contradiction emerges as \( |\alpha| > \sqrt{3}/7 \) is impossible for a two-carrier hole-doped system. Evaluation of \( \beta \) positions the system in the \( n_e > n_h \) region where \( \sigma_h \rightarrow 0 \).\(^{43}\) In fact \( \beta > \alpha^2 \) is not strictly satisfied. A plausible explanation for these contradictory results is that the Fermi surface structure contains more than two sheets. Since the Fermi surface is likely reconstructed by an incommensurate charge-density-wave order,\(^{35-37}\) a more complex multi-band structure is expected.\(^{38}\) On the electron doped side (\( n_h > n_e \)), by contrast, the Fermi surface is expected to fold around the antiferromagnetic zone boundary leading to a two-band structure\(^{33}\) (see Fig. 4a).

Quantum oscillation Hall experiments on \( \text{Pr}_1.95\text{Ce}_{0.05}\text{CuO}_4 \) (PCCO)\(^{39}\) and \( \text{Nd}_{1.95}\text{Ce}_{0.05}\text{CuO}_4 \) (NCCO)\(^{40,41}\) reveal a small hole pocket with \( n_h = 4.0 \times 10^{25} / \text{m}^3 \) giving \( R_H \ll R_c \). To compare to our model results, we use momentarily \( n_h \approx n_e \) and \( \sigma_e \approx \sigma_h \). We therefore have \( |\alpha| \ll 1 \) and evaluation of the magnetoresistance for both NCCO and PCCO\(^{30,42}\) yields \( \beta \approx 0.03 \) less than \( 3(1-\alpha^2)/4 \) as expected. These systems are thus not incompatible with a two-band model.

Next, we discuss our magnetoresistance experiments on \( \text{Ca}_3\text{Ru}_2\text{O}_7 \). Recent ARPES data suggest that \( \text{Ca}_3\text{Ru}_2\text{O}_7 \), at base temperature, is a semimetal with a single electron and two identical hole pockets\(^{33-35}\) (see Figure 4b). The electron pocket - corresponds to a carrier density \( n_e = 7.8 \times 10^{24} / \text{m}^3 \), consistent with the dominant quantum oscillation frequency.\(^{27}\) The electron Hall coefficient \( R_e = -1/\epsilon n_e \) is therefore known within the confidence provided by ARPES and quantum oscillation experiments. Our low magnetic-field magnetoresistance data permit extraction of \( R_H, \sigma \) and \( C \) as a function of temperature. At \( T = 10\ \text{K} \), we find \( \alpha = -0.21 \) and \( \beta = 0.85 \) and with that, all model parameters are now known: \( \sigma_h = 0.37\sigma = 0.6\sigma_e \), and \( |R_h/R_e| = 1.26 \) (see Fig. 3). The electron-hole carrier ratio is consistent with the low-temperature ARPES derived Fermi surface.\(^{43}\) Our results also imply that the electron carriers are more mobile \( \mu_e = 1.33\mu_h \) than the holes. This is consistent with the fact that the largest quantum oscillation amplitude\(^{27}\) is found for the frequency corresponding to the electron pocket in the ARPES experiment.\(^{43,44}\) So far, the two-band model seems to sensibly describe the magnetoresistance experiment. However, there are at least three problematic issues with the two-band model. (i) Even in the temperature range \( 2\ \text{K} - 30\ \text{K} \), \( \alpha \) and \( \beta \) vary in a fashion that is inconsistent with a constant \( n_h/n_e \). (ii) For intermediate temperatures negative magnetoresistance is observed. (iii) We used the low-field limit \( H \rightarrow 0 \) for our analysis. On top of this, higher order magnetic field terms can be derived. However, these higher-order terms are not large enough to account for the deviation from \( \rho_{xx} \propto H^2 \) under higher fields (Fig. 2d).

In what follows, we discuss possible reasons for these discrepancies. We start by stressing that the underlying assumption that carrier mobility is magnetic-field independent is not the sole culprit. Field dependent mobilities could explain (iii) but not (i) and (ii). We also point out that carrier density ratio \( n_h/n_e \) may, in fact, be temperature dependent. Recent density-functional-theory (DFT) calculations suggest a strong coupling between detailed lattice structure and a temperature dependent Coulomb interaction.\(^{45}\) Early neutron diffraction experiments indeed demonstrated the temperature dependence of ruthenium-oxygen bond angles.\(^{46}\) It is therefore not inconceivable that the chemical potential and even the low-energy electronic structure is temperature dependent. In fact, recent ARPES studies reported a two stage reconstruction with characteristic temperatures of \( T^* = 30\ \text{K} \) and \( T_s = 48\ \text{K} \).\(^{43,44}\) Since the negative magnetoresistance is found for \( T^* < T < T_s \), it is very likely linked to the inter-
mediated reconstructed electronic structure (see Figure 4c). ARPES experiments suggest that these Fermi surface consists for more than two sheets. Furthermore, out-of-plane resistivity indicate that a finite inter-layer interaction is still present. As such the electronic structure contains complexity beyond the most simple two-band model. In addition, our magnetization data (see Figure 2b) suggests the existence of weak ferromagnetism (on top of the antiferromagnetic coupling) below $T_s$. It is not uncommon that magnetic correlations generate a negative magnetoresistance that is comparable or larger than the orbital component. It is also not unusual that the ferromagnetic contribution is largest just below the ordering temperature. The negative magnetoresistance in Ca$_2$Ru$_2$O$_7$ therefore is likely linked both to the electronic structure and ferromagnetic properties.

6. Conclusions

In summary, we have carried out a magnetoresistance study of Ca$_2$Ru$_2$O$_7$. As a function of temperature, three different regimes are identified: weak positive magnetoresistance for $T > 48$ K, negative magnetoresistance for $30 < T < 48$ K and large positive magnetoresistance below 30 K. These characteristic temperatures are directly linked to reconstructions of the low-energy electronic structure. We analysed the low-temperature magnetoresistance within a two-band model. An analytical solution to the two-band model for conductivity is developed and expressed in terms of two dimensionless (but measurable) parameters. Whereas reasonable values of electron/hole carrier density and mobility is found for selected temperatures ($T \approx 10$ K), the two band model is not capturing the full magnetic field and temperature dependence. The most pronounced limitation of the model is its inability to explain negative magnetoresistance. On top of a two-stage electronic reconstruction, we argue that the Fermi surface structure – and with that the electron/hole carrier density – is temperature dependent. This in combination with ferromagnetism generates the complicated magnetoresistance in Ca$_2$Ru$_2$O$_7$.

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Appendix

In the main text, the dimensionless parameters $\alpha$ and $\beta$ are expressed in terms of $\sigma_n/\sigma = F(\alpha, \beta)$ and $R_\perp/R_\parallel = G(\alpha, \beta)$. – Equations (3) and (4). Solving with respect to $F(\alpha, \beta)$ and $G(\alpha, \beta)$ yields:

$$F(\alpha, \beta) = A_1 + \frac{A_2}{(A_3 + A_4)^{1/3}} + \frac{2^{2/3}}{6} (A_3 + A_4)^{1/3}, \quad (A.1)$$

and

$$G(\alpha, \beta) = \frac{\alpha + [1 - F(\alpha, \beta)]^2}{[F(\alpha, \beta)]^2}. \quad (A.2)$$

with

$$A_1 = 1 + \frac{2}{3} \alpha,$$

$$A_2 = \frac{\sqrt{2}}{3} (\alpha^2 - \beta),$$

$$A_3 = -[2\alpha^3 + 3\beta(2\alpha + 3)],$$

$$A_4 = \sqrt{3} \beta \sqrt{\frac{12\alpha^2(1 + \alpha)}{\beta} + (27 + 36\alpha + 8\alpha^2) + \frac{4\beta}{3}}. \quad (A.3)$$

It is now possible to express the longitudinal and transverse resistance in terms of $F$ and $G$:

$$\rho_{xx} = \frac{1 + F(1 + \frac{\mu_e}{\mu_H})^2}{\sigma + (1 - G)\mu_e H}, \quad (A.4)$$
For a semimetal with $\mathcal{G} = 1$ ($n_e = n_h$), Equations A-4 and A-5 simplify to $\Delta \rho(\mathcal{G} \rho(0) = \mu_e \mu_h \mathcal{B}^2$ and $\rho_{xy} = (1 - \mu_e / \mu_h) R$. Therefore, no saturation of magnetoresistance or Hall effect is expected. For $\mathcal{G} \neq 1$, the magnetoresistance should eventually saturate and non-linear $\rho_{xy}$ isothersms should occur in the high-field limit. As shown in Fig. 2d, we indeed observe saturation of the $\Delta \rho(\mathcal{G} \rho(0) \propto H^2$ and $\rho_{xy}$ as magnetic field strength is increased. In the same field range, the magnetization scales perfectly with $H$. Hence ferromagnetization as a source for the saturating magnetoresistance can thus be excluded. As we find $\mathcal{G}$ close to unity, this saturation effect is stronger than what can be explained by the two-carrier model.

It is possible to fit the entire field range, however the result is stronger than what can be explained by the two-carrier model.