Sign changes of the Hall coefficient in Sr$_2$RuO$_4$ reveal coherent-to-incoherent and elastic-to-inelastic crossovers

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The Hall coefficient $R_H$ of Sr$_2$RuO$_4$ exhibits an unusual non-monotonic temperature dependence with two sign reversals. We show that this behavior is the signature of two crossovers which are key to the physics of this material. The initial rise upon cooling from high temperatures is due to the strong temperature and orbital dependence of the inelastic scattering rates, signaling the gradual formation of coherent quasiparticles, whereas the eventual decrease at low temperatures is driven by the crossover from inelastic to impurity-dominated scattering. These qualitative conclusions are supported by a quantitative calculation of $R_H(T)$ using Boltzmann transport theory in combination with dynamical mean-field theory. Because it affects the topography of the Fermi surface sheets and strongly mixes their orbital character, spin-orbit coupling is found to be essential in explaining the behavior of the Hall coefficient.

Measuring the Hall coefficient $R_H$ is a standard way of characterizing charge carriers in a condensed matter system. For free carriers of a single type the Hall coefficient $R_H$ is simply given by the inverse of the density of carriers $n$ and their charge $e$. However, in complex materials with a Fermi surface (FS) composed of multiple sheets, interpreting $R_H$ can be more complicated when both electron-like and hole-like carriers are present. For instance, in the case of one hole-like and one electron-like FS sheet, the corresponding Hall coefficient is given by an average of $R_{H,e} < 0$ and $R_{H,h} > 0$:

$$R_H = \frac{\sigma_e^2 R_{H,e} + \sigma_h^2 R_{H,h}}{(\sigma_e + \sigma_h)^2},$$

weighted by the squares of the individual hole and electron conductivities, $\sigma_h$ and $\sigma_e$, respectively. Hence, the ratio of scattering rates between the two types of carriers enters in a key manner to determine both the overall sign and magnitude of the Hall coefficient.

The 4d transition metal oxide Sr$_2$RuO$_4$ is such a complex material: with low-energy bands built out of three Ru-$t_{2g}$ orbitals ($d_{xy}, d_{yz}, d_{zx}$) hybridized with O-$2p$ states, it has a FS comprising two electron-like sheets, $\beta$ and $\gamma$, and one hole-pocket, $\alpha$ [1–3]. And indeed, experiments [4–6] have observed a particularly interesting temperature dependence of $R_H$ in Sr$_2$RuO$_4$ (see Fig. 1): $R_H$ increases from a negative value of about $-1 \times 10^{-10} \text{m}^2\text{C}^{-1}$ at low temperatures [7], exhibits a sign change at 30 K (in the cleanest samples), reaches a positive maximum at about 80 K, changes sign a second time around 120 K and eventually saturates to a slightly negative value for $T > 200$ K.

Obviously, this rich temperature dependence is a signature of the multi-carrier nature of Sr$_2$RuO$_4$, as realized early on in Ref. [4], which considered a Drude model with two types of carriers. This was refined later in several works [8–10] using Boltzmann transport theory calculations for tight-binding models assuming scattering rates $1/\tau_\nu = A_\nu + B_\nu T^2$ for the different FS sheets $\nu = \{\alpha, \beta, \gamma\}$, with adjustable parameters $A_\nu$ and $B_\nu$.

The overall take-home message of these phenomenological models is that $R_H$ is highly sensitive to the precise details of the FS sheets and also to the temperature and sheet dependence of the scattering rates.

Another remarkable experimental finding provides insight in interpreting the temperature dependence of $R_H$ [6]: Adding small amounts of Al impurities has a dramatic impact on the intermediate temperature regime such that $R_H$ no longer turns positive and instead increases monotonically from the low-T to the high-T limit, as indicated by dotted lines in Fig. 1. Arguably, the similarity of the low-T values of $R_H$ for different impurity concentrations provides evidence that the elastic-scattering regime has been reached where $R_H$ is mainly determined by FS properties (see also Ref. [5]). In contrast, the temperature dependence itself must be due to inelastic scattering, possibly associated with electronic correlations [6].

In this letter, we address the rich temperature dependence of $R_H$ in Sr$_2$RuO$_4$ and provide a clear interpretation of its physical meaning, as well as an explicit calculation of $R_H(T)$ beyond phenomenological models. We show that the two sign-changes of $R_H(T)$ in clean samples are the signatures of two important crossovers in
the physics of this material. The increase of $R_H$ upon cooling from high temperature signals the gradual formation of coherent quasiparticles, which is associated with a strong temperature dependence of the ratio of inelastic scattering rates between the $xy$ and $xz/yz$ orbitals. At low temperatures the decrease of $R_H$ is due to the crossover from inelastic to impurity-dominated scattering. This qualitative picture is supported by a quantitative calculation of $R_H(T)$ using Boltzmann transport theory in combination with dynamical mean-field theory (DMFT) [11], taking into account the electronic structure of the material. The spin-orbit coupling (SOC) is found to play a key role [12; 13], because it has a strong influence on the topography of the FS and also controls the manner in which the scattering rates associated with the different orbitals combine into $k$-dependent quasiparticle scattering rates at a given point on the FS [12–14]. Moreover, recent works showed that electronic correlations enhance the effective value of the SOC by nearly a factor of two [15–17], which is crucial to obtain a perfect agreement with photoemission spectroscopy [14]. We believe that the interplay between FS topography, SOC, and strongly orbital-dependent scattering rates has wide relevance beyond the specific case of Sr$_2$RuO$_4$ and that our analysis provides insights into the physical significance of the temperature dependence of $R_H$ often observed in materials with strong electronic correlations and a complex FS [18].

We begin by discussing the key importance of the orbital dependence of scattering rates irrespective of the microscopic details of the underlying scattering mechanisms, which will be addressed at a later stage. We use Boltzmann transport theory to calculate $R_H$, as defined and implemented in the BoltzTraP software package [19–21]. The used Hamiltonian is obtained from a Wannier function construction for the three Ru-4$d$ $t_{2g}$ orbitals, as detailed in Ref. [14]. We compare calculations with and without SOC, and note that in the following all results labeled with ‘SOC’ actually take the correlation-enhanced effective SOC into account [14; 16; 17; 22]. We assign scattering rates $\eta_{xy}, \eta_{xz} = \eta_{yz}$ [23] to a given orbital. Then, these scattering rates are converted into band and $k$-dependent scattering rates using the overlap of the orbital wave-function with the Bloch wave-function [14; 24]:

$$\eta_m(k) = \sum_m |\langle \psi_m^k | \chi_m \rangle|^2 \eta_m$$

with $m = \{xy, xz, yz\}$. $R_H$ only depends on scattering rates through their ratio $\xi = \eta_{xy}/\eta_{xz/yz}$ and not their absolute magnitude; a point we verified in our calculations. This also implies that within the constant isotropic scattering rate approximation, i.e. $\xi = 1$, the full temperature dependence of $R_H$ cannot be explained.

The calculated $R_H$ as a function of the scattering rate ratio $\xi$ is displayed in Fig. 2. Without SOC $R_H$ remains negative for all values of $\xi$ and approaches zero as $\xi \to 1$. In this limit, the $\gamma$ sheet drops out and the contributions of the hole-like $\alpha$ sheet and electron-like $\beta$ sheet compensate each other. This means that it is not possible to explain the positive value of $R_H$ observed experimentally in clean samples for $T_1 < T < T_2$ (Fig. 1) without taking SOC into account. With SOC we observe a very different behavior of $R_H(\xi)$; it turns from negative to positive at $\xi \simeq 2.6$. This is a result of two effects (Fig. 2, inset): First, SOC changes the shape and size of the FS sheets, and secondly, it induces a mixing between different orbital characters, which varies for each point on the FS. From the calculated dependence of $R_H(\xi)$ in the presence of SOC we deduce that agreement with experiments would require $\xi$ to be smaller than 2.6 at high temperatures, increase above this value at $\sim T_2$ and then decrease again to reach a value close to unity at low temperatures.

We turn now to microscopic calculations by first consider inelastic electron-electron scattering ratios calculated with DMFT. These calculations consider the $t_{2g}$ subspace of states with Hubbard-Kanamori interactions of $U = 2.3$ eV and $J = 0.4$ eV [25; 26]. The calculated $\xi(T)$ from inelastic scattering only is displayed in Fig. 3 (a). In agreement with previous studies [25; 27], we find that the $xy$ orbital is less coherent than $xz/yz$ at all temperatures and $\eta_{xy} > \eta_{xz/yz}$. In Sr$_2$RuO$_4$ the crossover from the low-T coherent Fermi liquid regime with $\eta \sim T^2$ to an incoherent regime with a quasilinear temperature dependence of the scattering rate is well-documented [25; 28] and also manifested in deviations of the resistivity from a low-temperature quadratic behavior to a linear one [29]. Importantly, this coherence-to-incoherence crossover as well as the corresponding coherence scales are strongly orbital dependent. When approaching the Fermi liquid regime ($T_{FL} = 25$K [29–31]) the scattering rate ratio reaches a value as large as
\( \xi^{FL} \sim 3 \) (Fig. 3 (a)), but decreases rapidly upon heating with \( \xi = 1.8 \) at 300 K. For an even higher temperature of 500 K (not shown) the ratio is hardly different (\( \xi = 1.6 \)), which indicates that a fully incoherent regime has been reached.

Connecting these results to the discussion of Fig. 2 above, the temperature dependence of \( \xi \) directly translates into that of \( R_H \), as shown in Fig. 3 (b). Like in experiments, \( R_H \) is negative at high temperatures, but when the temperature is lowered it increases and crosses zero at 110 K. This demonstrates that electronic correlations are indeed able to turn \( R_H \) positive and suggests the following physical picture: The electronic transport in \( \text{Sr}_2\text{RuO}_4 \) crosses from a regime governed by incoherent electrons at high temperatures, connected to a weaker orbital dependence of scattering rates and a negative \( R_H \), over to a coherent Fermi liquid regime, with a stronger orbital dependence and positive \( R_H \). The resulting sign change at 110 K can be seen as a direct consequence of this coherence-to-incoherence crossover. We emphasize that this sign change is only observed when SOC is taken into account. Without SOC, \( R_H \) is purely negative and shows only a weak temperature dependence (Fig. 3 (b), dashed line).

When moving along the FS from \( \Gamma-\text{M} \) (\( \theta = 0^\circ \)) to \( \Gamma-\text{X} \) (\( \theta = 45^\circ \)), the mixing of the orbital character induced by SOC, leads to angular-dependent scattering rates \( \eta_\alpha(\theta) \) (Fig. 3 (c)). At \( \theta = 0^\circ \) the ratio of scattering rates between the \( \gamma \) and \( \beta \) sheets is large, because these bands still have mainly \( xy \) and \( xz/yz \) character, respectively (Fig. 2, inset). As expected from Fig. 3 (a), this sheet dependence decreases with increasing temperature. On the other hand, at \( \theta = 45^\circ \) the ratio is small, due to a very similar orbital composition of the \( \gamma \) and \( \beta \) sheets. The \( \alpha \) pocket (being almost entirely \( xz/yz \)) has the lowest scattering rate and turns \( R_H \) positive when \( \xi \) becomes large enough at low temperatures. To shed more light on the interplay of the individual FS sheets we can phenomenologically assigning constant scattering rates to each FS sheet, as shown in Fig. 3 (d). We see that for \( R_H \) to be positive a necessary condition is \( \eta_\beta > \eta_\alpha \). This again demonstrates the importance of SOC, because without SOC the \( \alpha \) and \( \beta \) sheets have entirely \( xz/yz \) orbital character, and thus \( \eta_\beta > \eta_\alpha \). Should one make this assumption also in the presence of SOC, it would not result in \( R_H > 0 \) for any ratio \( \eta_\beta/\eta_\alpha \) (Fig. 3 (d), dashed line). Assigning a scattering rate to a specific orbital and taking into account the orbital composition of each quasiparticle state on the FS seems to be the more appropriate picture, as indeed done within the DMFT framework above. This key insight, which has been recently supported by angle-resolved photoemission spectroscopy [14], is likely to have a large degree of generality in other multi-orbital materials.

Considering inelastic scattering only would yield a positive \( R_H \) at even lower temperatures deep in the Fermi liquid regime. However, at such low temperatures elastic scattering is expected to dominate over inelastic scattering. The extracted DMFT scattering rates at 29 K with 5.5 meV for the \( xy \) and 1.9 meV for the \( xz/yz \) orbitals are of the order of the impurity scattering for ‘clean’ samples with residual resistivities of \( \sim 0.5 \mu\Omega\text{cm} \). Therefore, we add a constant elastic scattering \( \eta^{\text{el}} \) to the orbital-
The corresponding crossover scale is obtained by inelastic scattering at higher temperatures. Matching the crossover between the regime dominated by elastic scattering with inelastic scattering shapes the overall temperature dependence of the Fermi liquid coherence scale $R_{FL}$ at $29$ K. The inset shows the experimentally measured $R_H$ from Ref. [6] for samples with different residual resistivities $\rho$ obtained by introducing small amounts of Al impurities.

With increasing temperature the influence of elastic scattering fades away and the precise interplay with the inelastic scattering rates at 29 K. For small enough $\eta_{el}$ we observe a second zero crossing of $R_H(T)$ and a regime with $R_H < 0$ at low temperatures, which is consistent with $R_H(T)$ depicted in Fig. 1. For $T \to 0$ the fully elastic scattering regime is reached, and thus $R_H$ is not influenced by the magnitude of the (isotropic) scattering rate, but rather by the shape of the FS only. This regime corresponds to $\xi = 1$ in Fig. 2, for which we obtain $R_H = -0.94 \times 10^{-10}$ m$^3$ C$^{-1}$, in good quantitative agreement with experiments [7]. With increasing temperature the influence of elastic scattering fades away and the precise interplay with the inelastic scattering shapes the overall temperature dependence of $R_H$. Hence, we see that also the low-temperature zero crossing has a simple physical interpretation: it signals the crossover between the regime dominated by elastic scattering at low temperatures and the regime dominated by inelastic scattering at higher temperatures. Matching the two terms in the scattering rate, a simple estimate of the corresponding crossover scale is $T_1 \sim \sqrt{\eta_{el}/A_{xy}} \sim \sqrt{\eta_{el}} T_{FL}$. This scale obviously depends on the elastic scattering rate, and coincides approximately with the Fermi liquid coherence scale $T_{FL}$ only for the cleanest samples reported in which $\eta_{el} \sim T_{FL}$. For even cleaner samples we predict $T_1 < T_{FL}$.

On the contrary, for larger $\eta_{el}$ we find that $R_H(T)$ ceases to exhibit any zero crossing and is negative in the whole temperature range. Only in very clean samples is the crossover from elastic to inelastic scattering sufficiently separated in energy from the coherent-to-incoherent crossover to reveal the sign changes of $R_H$. This is further substantiated by experimental Hall measurements for crystals where the residual resistivity was altered by introducing different amounts of Al impurities, cf. the dependence of $R_H$ on $\eta_{el}$ in Fig. 4 and the inset with experimental data from Ref. [6].

In the high-$T$ limit we obtain a value of $R_H$ which is more negative than the one reported in experiments [4–6]. Within Boltzmann transport theory this would imply that a larger ratio $\eta_{xy}/\eta_{zz/yz}$ is needed. Likewise, resistivities are significantly underestimated in DMFT transport calculations for $T > 300$ K in this material [27]. A possible explanation is that other sources of inelastic scattering, e.g. electron-phonon scattering, could play an important role in the high-$T$ regime. We emphasize, however that all experimental evidence points towards negligible magnetic contribution (due to processes like skew scattering) and a standard orbital-dominated Hall effect in Sr$_2$RuO$_4$ [5; 9; 32].

Summarizing, we have provided a qualitative interpretation and quantitative calculations of the complex temperature dependence of the Hall coefficient in Sr$_2$RuO$_4$, which involves the interplay of elastic impurity scattering and inelastic electron-electron scattering. A rather similar non-monotonic temperature dependence, although with purely positive Hall coefficient, has been experimentally observed in Sr$_3$Ru$_2$O$_7$ [33]. At a broad level, the Hall effect has been shown to be sensitive to the orbital content of quasiparticle states along different sheets of the Fermi surface, and as such is likely to be important in understanding many other multi-orbital materials in the presence of spin-orbit coupling. We note that recently interest has been devoted to the theoretical descriptions of the Hall effect in strongly-correlated multi-orbital systems beyond Boltzmann transport theory [34–36].

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We use a 46 x 46 x 46 \( k \)-grid, which is interpolated on a 5 times denser grid with BoltzTraP [19; 20]. Inter-band transitions are not considered in BoltzTraP, but we verified with Kubo transport calculations of the conductivity \( \sigma_{xx} \) (TRIQS/DFTTools [37]) that these are negligible.

Electronic correlations lead to approximately frequency-independent off-diagonal elements of the self-energy [17], which we absorb into a static effective SOC strength of \( \lambda_{eff} = 200 \text{meV} \) [14].

\( \eta_{xx} = \eta_{xx} \), due to the tetragonal crystal structure.

The (neglected) off-diagonal elements \( \eta_{\nu\nu'}(k) \) were shown to be small [14].

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