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Extraordinary Hall effect in $SrRuO_3$

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We have measured the Hall effect in thin films of the itinerant ferromagnet $SrRuO_3$ as a function of temperature (1.8 K–300 K) and field (0–8 T). We find that the extraordinary Hall coefficient R_s changes sign and can be described as $R_s \approx cT^5 + b\rho^2$ where T is temperature and ρ is resistivity. We discuss possible interpretations of this result.

The Hall resistivity (ρ_H) in magnetic metals is commonly given by $\rho_H = R_0 B + R_s \mu_0 M$, where B and M are the magnetic induction and the magnetization, respectively. The term ($R_0 B$) is related to the effect of B on the trajectories of the charge carriers. It is present in all metals and R_0 is known as the regular Hall coefficient. The term ($R_s \mu_0 M$) is related to the effect of magnetic moments on the charge carriers trajectories. It is present only in magnetic metals and R_s is known as the extraordinary (or anomalous) Hall constant. While the regular Hall effect is by now well understood, the understanding of the extraordinary Hall effect (EHE) is far from being complete despite considerable theoretical[1] and experimental[2] efforts. This is partly due to the difficulty to distinguish between the regular and extraordinary contributions to the Hall effect when the magnetization induces a large magnetic field and/or large fields are needed to magnetize the sample (due to formation of magnetic domains). In the following we present EHE data of $SrRuO_3$ for which the uncertainty in measuring the EHE is minimal and compare them to existing theoretical models and experimental observations.

The common theoretical view is that

$$R_s = a\rho + b\rho^2, \tag{1}$$

where ρ is the longitudinal resistivity and a and b are constants. The linear term in ρ is attributed to asymmetric scattering of charge carriers, a process which derives from the *classical* Boltzmann equation. On the other hand, the quadratic term in ρ is attributed to asymmetric side jumps which is a purely *quantum* scattering process. While it is preferable to test $R_s(\rho)$ when ρ is dominated by point defects and is modified by increasing the density of impurities (since in this case a single type of scattering is involved), numerous recent reports have examined $R_s(\rho)$ when the change in ρ was achieved by changing the temperature.[3,4] The experiments (both recent and previous ones) seem to support the claim of two distinct processes being responsible for the EHE: $R_s(\rho)$ of low-resistivity magnetic metals (e.g., dilute alloys at low temperatures) is usually linear in ρ while $R_s(\rho)$ of high-resistivity compounds (either due to disorder or high temperature) is usually quadratic in ρ.

Here we present measurements of the EHE in $SrRuO_3$ as a function of temperature. We find that at low temperatures R_s is negative and proportional to ρ^2. As temperature increases, the slope of dR_s/dT changes sign and close to T_c, R_s turns positive. The change of sign clearly indicates that in addition to the negative quadratic term there is also a positive term. However, we show below that this term is not linear in ρ; namely, Eq. (1) does not fit our data. On the other hand, a reasonable fit for R_s is obtained with $R_s = cT^5 + b\rho^2$. In the following we discuss possible implications of this result.

The orthorhombic perovskite $SrRuO_3$ ($a \sim 5.53$ Å, $b \sim 5.57$ Å, $c \sim 7.85$ Å) is an itinerant ferromagnet with Curie temperature (T_c) of about 150 K for films and more than 160 K for bulk. Our measurements were done on thin films of $SrRuO_3$ grown on slightly-miscut ($\sim 2°$) substrates of $SrTiO_3$. The $SrRuO_3$ film grows on such substrates as an untwinned single-crystal film with its [001] ([$\bar{1}$10]) direction in the film plane and perpendicular to (along) the miscut direction. The saturated magnetic moment of $SrRuO_3$ is $\sim 1.4 \mu_B$ per ruthenium and, due to the uniaxial magnetocrystalline anisotropy, it lies approximately along the [010] direction which in our films is at 45° out of the plane of the film (bisecting the 90° angle of the unit-cell terraces of the miscut $SrTiO_3$).[5] For this study we have used high-quality single-crystal films grown by reactive electron beam coevaporation with residual resistivities as low as 4.6 $\mu\Omega$ cm (corresponding to a resistivity ratio between $T = 300$ K and $T = 1.8$ K of ~ 45) and thicknesses between 800 and 2000 Å. The magnetization of the films was measured with a Quantum Design SQUID magnetometer and subsequently the films were patterned to allow for resistivity and Hall effect measurements with the current along the [001] and [$\bar{1}$10] directions in each film.

To extract R_s we measured ρ_H as a function of field at different temperatures (see Fig. 1). The Hall resistivity comprises both the regular and the extraordinary part. However, two important properties of $SrRuO_3$ allow for minimal uncertainty in the identification of the EHE part below T_c. First, after fully aligning the magnetization of the sample by applying high magnetic fields, there is no nucleation of reversed magnetization regions when the field is set to zero.[6]

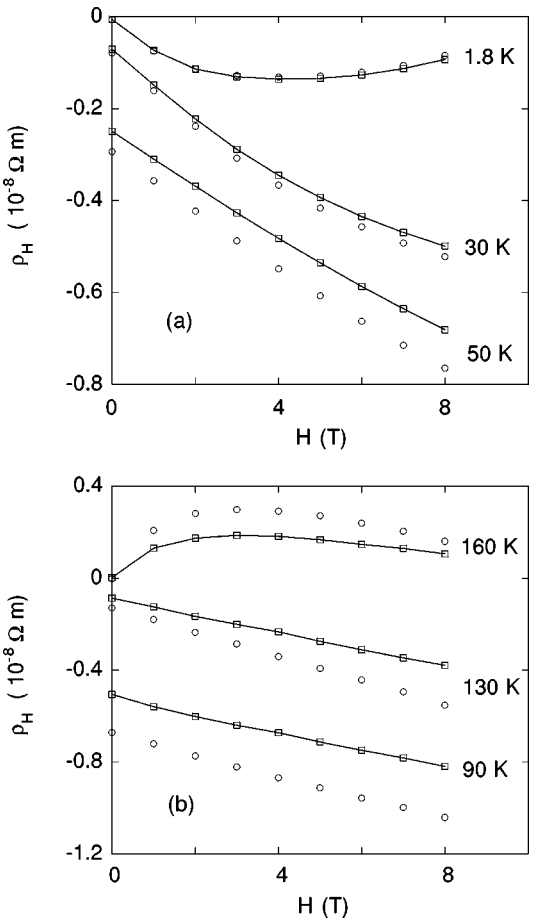

FIG. 1. (a) Hall resistivity as a function of field at $T=1.8$, 30, and 50 K. The field is perpendicular to the film plane and the current is along the [001] direction (connected open squares) and the $[\bar{1}10]$ direction (open circles). (b) Hall resistivity as a function of field at $T=90$, 130, and 160 K. The field is perpendicular to the film plane and the current is along the [001] direction (connected open squares) and the $[\bar{1}10]$ direction (open circles).

Therefore, in a remnant state we measure the full contribution of the spontaneous magnetization to the Hall effect. Second, the self field created by the remnant M is less than ~ 2 KOe which induces a small regular Hall effect whose contribution to the total Hall effect at zero applied field can be generally neglected. As a result of these two properties we may reliably identify the zero field Hall effect with the EHE. The interpretation of the Hall effect becomes more complicated when an external field is applied since two sources affect ρ_H: (a) the regular Hall effect and (b) the change in the EHE due to the change in M. For this reason, the determination of R_0 requires the use of a field range where the change in the EHE is negligible compared to changes in the regular Hall effect. Based on magnetization measurements we assume that at $T<140$ K most of the variation in ρ_H for fields above $H=4$ T is related to the regular Hall effect.

Before turning to the EHE we examine the regular Hall effect mainly for its role in analyzing the EHE (see Fig. 1). We see that in the temperature interval 60 K$<H<$140 K and at fields of 4 $T<$T$<$8 T $\rho_H(H)$ is quite linear. Attributing this behavior to the regular Hall effect we estimate R_0 and using the relation $R_0=1/ne$ we find $n\sim 1.5 \times 10^{22}$ 1/cm^3 which corresponds to approximately one charge carrier per ruthenium. Below 60 K, ρ_H becomes increasingly nonlinear at high fields; see, e.g., the behavior of ρ_H at $T=1.8$ K [Fig. 1(a)] where the slope of ρ_H turns from negative to positive at $H>4$ T. This behavior of $\rho(H)$ may be quite confusing since it is reminiscent of the behavior of ρ_H in other itinerant ferromagnets for which the change of slope occurs when the change in the regular Hall effect exceeds the change in the EHE. In such cases the high field part of $\rho(H)$ is extrapolated to $H=0$ to estimate the EHE. This, however, is not the case here since it would imply that at $T=1.8$ K the EHE at saturation is more than 30 times larger than its magnitude in remnant state at $H=0$ while we know that $1-M/M_s<0.001$, where M is the remnant magnetization and M_s is the saturation magnetization. Therefore, contrary to previous conclusions,[7] we find that $R_0(H=0)$ does not change sign. It remains electronlike in the low temperatures limit while the sign change in the slope of ρ_H reflects a sign change of dR_0/dH. Such a sign change is commonly related to low-field high-field crossover which occurs when $\omega_c\tau\sim 1$. Here $\omega_c=(eH/m_Hc)$ is the cyclotron frequency (m_H is the cyclotron mass) and τ is the scattering time. When the temperature is increased, τ decreases and consequently the field at which $\omega_c\tau\sim 1$ is satisfied increases. Therefore, at, e.g., 20 K the crossover field is greater than our maximum field of 8 T; nevertheless, the effect of the incipient crossover on ρ_H is clearly observed. The crossover field allows us to estimate the scattering time, $\tau=1/\omega_c=5.68\times 10^{-12}\times(m_H/m_eH)$ where H is the crossover field in teslas. At $T=1.8$ K the crossover field is ~ 4 T and using the calculated Fermi velocity of $v_F\sim 2\times 10^7$ cm/sec (Ref. 8) we find that the mean free path l is $l=v_F\tau\sim 2.84(m_H/m_e)\times 10^{-5}$ cm. On the other hand, the estimated average mean free path which corresponds to a resistivity of 4.6 $\mu\Omega$ cm is about 5×10^{-6} cm. Therefore, our results imply that $(m_H/m_e)\sim 0.2$ (while $m^*/m_e\sim 3.7$).[8] This very small m_H is consistent with quantum oscillations measurements performed on similar films which showed that the main signal corresponds to a cyclotron mass of 0.2 electron masses.[9]

Having identified the regular Hall effect we turn to examine R_s. In Fig. 2(a) we show the extraordinary Hall resistivity ($R_s\mu_0M$) at $H=0$ as a function of temperature with the current along the [001] and $[\bar{1}10]$ directions. We see that $R_s\mu_0M$ is small and negative at low temperatures. As temperature increases, $R_s\mu_0M$ becomes more negative reaching a minimum at about 90 K, after which it increases and crosses zero at about 130 K. Since we show the EHE at $H=0$, it vanishes at $T>T_c$. We note that there is a difference in the EHE for the two current directions. This behavior may be related to the different angles between the currents and the magnetic moment which is $\sim 45°$ for the current in the $[\bar{1}10]$ direction and $\sim 90°$ for the current in the [001] direction. It is interesting to note that the anisotropy in the EHE is much larger than that observed in the longitudinal resistivity. It could be either because the EHE is more sensitive to the anisotropy in the conducting states in the two directions or because the EHE is sensitive to the angle between the current and the magnetic moment and does not depend only on the perpendicular component.

To test the applicability of Eq. (1) we plot in Fig. 2(b) R_s (obtained by dividing the extraordinary Hall resistivity by

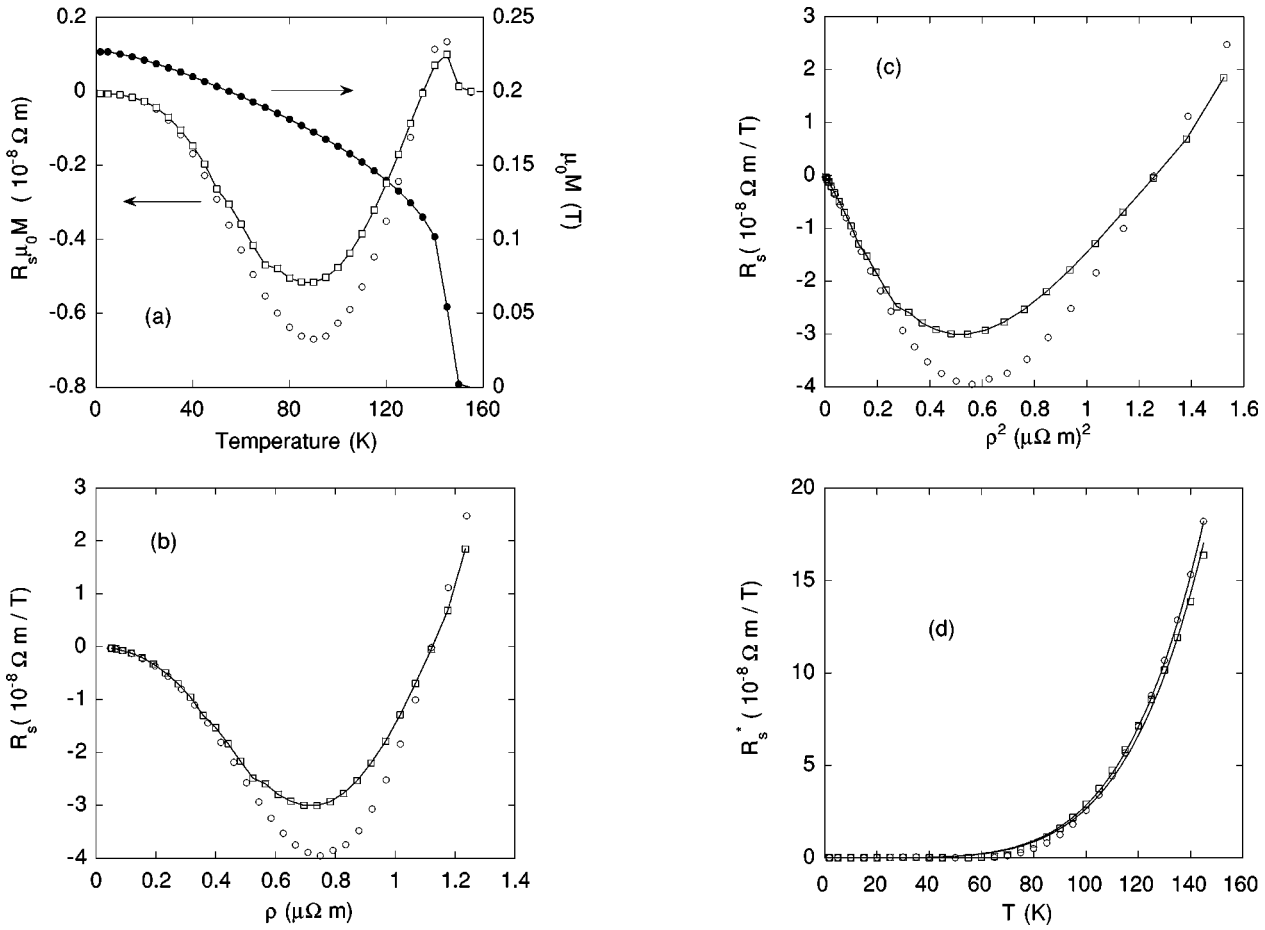

FIG. 2. The EHE for current along the [001] direction (connected open squares) and in the $[\bar{1}10]$ direction (open circles). (a) The extraordinary part of the Hall resistivity as a function of temperature. Also shown $\mu_0 M$ (full circles) as a function of temperature where M is the component of the remnant magnetization that is perpendicular to the plane of the film. (b) The coefficient of the EHE as a function of resistivity. (c) The coefficient of the EHE as a function of resistivity square. (d) The coefficient of the EHE without the part quadratic in resistivity as a function of temperature.

$\mu_0 M$) as a function of the resistivity ρ. We see that Eq. (1) cannot fit our data. The theoretical prediction is for a parabola whereas $R_s(\rho)$ is clearly not. Nevertheless, we observe that at low temperatures we can fit $R_s(\rho)$ by $R_s = b\rho^2$ [see Fig. 2(c)]. Assuming that the low temperature behavior $R_s = b\rho^2$ is due to side jumps we calculate its magnitude. The theoretical prediction is that $\Delta y = (b\mu_0 M_s \hbar k_F)/(2ne^2)$ where M_s is the saturation magnetism and n is the carrier density. Substituting $b \sim -1 \times 10^{-5}$, $\mu_0 M_s = 0.2$ T, $\hbar k_F \sim 7 \times 10^{-25}$ kg m/sec, and $n = 1.5 \times 10^{22}$ 1/cm^3 we obtain that $\Delta y \sim 0.2 \times 10^{-10}$ m. This result is within the common range of Δy's (10^{-11}–10^{-10} m) obtained for transition metal alloys.

Taking the reasonable value of Δy as an indication that there is a $b\rho^2$ contribution to R_s, we want to identify the additional contribution (beside the side jumps) to the EHE. For that we subtract from R_s the $b\rho^2$ contribution and examine $R_s^* = R_s - b\rho^2$. R_s^* is not linear in ρ as expected; therefore, we want to explore whether R_s^* is proportional just to part of the scattering events. To identify the remaining contribution we plot R_s^* as a function of T [Fig. 2(d)]. The simplest fit is in the form of $R_s^* \sim T^n$. Trying it, we find a reasonable fit with $n = 5$.

The ρ^2 term in R_s is considered more robust because it is generally believed that the side-jump mechanism is insensitive to the scattering mechanism;[1] hence, the relation is expected to hold even when several scattering processes contribute to the resistivity. It is not clear whether this is also the case for skew scattering.[1] Therefore, one way of analyzing our results is to identify the part of the resistivity that could give rise to a T^5 dependence. A possible candidate could be phonon scattering, since this is the phonon resistivity for $T \ll \theta_D$ where θ_D is the Debye temperature. However, θ_D of $SrRuO_3$ is ~368 K (Ref. 8) and the T^5 dependence is not expected to persist up to close to 150 K. It is more likely that T^5 would reflect some magnetic scattering; however, we are not aware of a magnetic scattering mechanism that will yield such a temperature dependence. Alternatively, it is possible that the difficulty in fitting the data is an anomaly of $SrRuO_3$. This compound exhibits various transport anomalies attributed to effects of strong electron correlations.[10] Therefore, it may be that a different theoretical treatment is required to calculate the anomalous Hall effect in this compound. Recently, it was reported that the EHE of manganites, which are strongly electron-correlated metals (below T_c) whose resistivity is dominated by magnetic disorder, cannot be fit with Eq. (1);[4] however, the data were fitted quite well with

new theoretical predictions[11] that took into consideration the particular transport properties of these compounds. It is possible that a similar treatment is required also in the case of $SrRuO_3$ whose EHE data cannot be fitted either with Eq. (1), or with the predictions for the manganites.

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