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Exotic physical properties in metallic perovskite LaRuO₃: a smoking gun for Hund metal

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The Hund's coupling rule is of fundamental importance in atomic physics. It also plays a key role in determination of physical properties in strongly correlated electron systems where the Fermi energy lies in multiple bands. We report a thorough investigation of a rarely studied 4d perovskite oxide LaRuO₃ by a suite of measurements including transport, magnetization, specific heat, thermoelectric powder, thermal conductivity, and neutron and X-ray diffraction. Our results are consistent with theoretical predictions, which make LaRuO₃ an exemplary Hund metal.

The Hartree-Fock (HF) approximation lays the foundation for treating many-body systems with single-body theory. The approach incorporates Coulomb interactions and spin-spin correlations between electrons at the mean field level and each electron is assigned the same spin that is delocalized in a short radius sphere in real space. In strongly correlated systems, the Coulomb interaction increases the probability for electrons to stay in the atomic orbitals in the Bloch waves. The real space confinement for electrons leads to a gap between the occupied and empty bands, *i.e.* the Mott transition. In a system where the Fermi energy crosses the bands arising from multiple orbitals, the Hund's coupling rule from atomic physics must be brought into the picture, **which leads to a Hund metal**. In this case, Hund's coupling regulates the magnitude of the spins, competes with the interatomic spin-spin interaction, and **reduces** the quasiparticle lifetime. **A characteristic and measurable property for a Hund metal is the thermally driven transition from coherent to incoherent state in which spins become localized.**^{1 2 3} The Hund metal has drawn attention since 2008. The model has been applied to rationalize physical properties observed in iron pnictide superconductors KFe_2As_2 ⁴ and Sr_2RuO_4 .^{5 6 7 8 9 7} **Whereas the transition of the transport property from the Fermi-liquid to a non-Fermi liquid behavior in KFe_2As_2 and the thermally-driven disappearing of the quasiparticle state from ARPES in Sr_2RuO_4 have reported,**^{6,7} the key feature of localized spin has been generally missing in these materials.^{10 11 12} In this Letter, we report a comprehensive set of measurements on the rarely studied metallic perovskite LaRuO_3 . An extremely strong intra-atomic interaction in the oxide enhances the Hund's coupling and ensures that LaRuO_3 represents an extreme case of a Hund metal.

In comparison with the well-known ferromagnetic metal $\text{SrRu}^{4+}\text{O}_3$,^{13 14 15 16 17 18 19 20 21 22 23 24 25} the perovskite LaRuO_3 has been rarely studied since Ru^{3+} is metastable in oxides. LaRuO_3 is the only member of the perovskite family RRuO_3 that can be synthesized at ambient pressure.^{26 27 28} High-pressure synthesis is needed for obtaining the entire family of polycrystalline RRuO_3 samples.^{29 30 31} By using the spark plasma sintering, we demonstrated the high density RRuO_3 pellet samples can be synthesized for $\text{R}=\text{La}$ and Pr . The resistivity of the LaRuO_3 sample is lower than all results reported in the literature. The detailed information about the sample synthesis and the characterization is provided in Supplemental Materials (SM).³² These oxides crystallize in the orthorhombic perovskite structure with the space group $Pnma$. The structural distortion originates from the cooperative octahedral-site rotations which bend the Ru-O-Ru bond angle ψ from 180° . As a direct measurement of the orthorhombic distortion, **the separation**

between lattice parameter a and c increases with increasing of the Pr doping x in $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$, see Fig.S2 in SM. By tuning the rare earth substitution, a reduction of electronic bandwidth in $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$ which is proportional to $\cos \psi$, leads to an evolution from metallic LaRuO_3 to an insulating PrRuO_3 .²⁹ The cell volume decreases gradually with increasing x , which resembles closely the cell volume change in the diamagnetic insulators $\text{La}_{1-x}\text{Pr}_x\text{GaO}_3$, see Fig.S2 in SM. This observation is in sharp contrast to the first-order Mott transition found in RNiO_3 .^{33 34} Moreover, no magnetic transition has been detected to the lowest temperature in the crossover from metallic LaRuO_3 to insulator PrRuO_3 , see the results of neutron diffraction and specific heat in SM.

Understanding the magnetic susceptibility χ is the first step for elucidating the physics in LaRuO_3 . Fig.1(a) shows the temperature dependence of χ^{-1} for the series of $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$. Fitting $\chi^{-1}(T)$ to a Curie-Weiss law seems to work reasonably well for $T > 100$ K. However, a large Weiss constant $\theta = -182.1$ K, a larger $\mu_{\text{eff}} = 2.556 \mu_{\text{B}}$ than the spin-only value, and a non-linear $\chi^{-1}(T)$ below 100 K make the practice questionable. Given that the spin-orbit coupling (SOC) is strong for the 4d electrons and the orbital angular momentum is not quenched in the low spin $\text{Ru}^{3+}: t_{2g}^5$, the influence of SOC must be considered. The Curie-Weiss law is the extreme case in the general formula of magnetic susceptibility for a negligible SOC.^{35 36} By working on a Hamiltonian for isolated magnetic ions, which includes single electron, spin-orbit coupling, and Zeeman effect contributions, Kotani³⁷ has derived an analytical expression for the magnetic susceptibility of compounds containing isolated 4d or 5d magnetic ions. In the Curie law expression, $\chi(T) = C/T$, the $\mu_{\text{eff}} \approx \sqrt{8C}$ becomes temperature-dependent. Kotani's modified expression for the effective magnetic moment in the system with $n=5$ is:

$$(1) \quad \mu_{\text{eff}}^2 = \frac{3x+8-8e^{-\frac{3}{2}x}}{x(1+e^{-\frac{3}{2}x})}, \text{ where } x=\lambda/kT \text{ and } \lambda \text{ is the spin-orbit coupling.}$$

The $\mu_{\text{eff}}(T)$ of LaRuO_3 shown in Fig.1(b) becomes temperature dependent below 100 K. Over a broad range of temperature, the $\mu_{\text{eff}} = 2.556 \mu_{\text{B}}$ of LaRuO_3 is clearly larger than the spin-only value for a d^5 low spin system. However, $\mu_{\text{eff}}(T)$ of LaRuO_3 matches the expression in Eq (1) for a d^5 system by Kotani stunningly well. Kotani's theory has been applied to successfully describe the magnetic susceptibility in insulators with isolated 4d or 5d magnetic ions.^{38 39} It is surprising

that the theory works well in metallic LaRuO₃. The best fitting of the experimental μ_{eff} at $T > 30$ K to the Kotani's theory is obtained with $\lambda = 0.015$ eV, which is much smaller than $\lambda = 0.07$ eV for insulators with isolated Ru³⁺.³⁹ The larger radial extension of the wavefunction reduces the atomic SOC parameter λ by a multiplicative fraction $k_c < 1$ (i.e. $k_c \lambda L \cdot S$).⁴⁰ The μ_{eff} predicted by the Kotani theory decreases with decreases temperature and it reaches $1.73 \mu_B$ at 0 K. The μ_{eff} of LaRuO₃ deviates from the theoretical curve below ~ 30 K. Since spins in LaRuO₃ do not order at low temperatures down to 1.5 K (see Fig.S4 in SM), the deviation of $\mu_{\text{eff}}(T)$ from Kotani's theory at $T < 30$ K may indicate a change of the electronic structure. This observation will be further discussed in connection with anomalies found in other physical properties.

The Pr substitution in La_{1-x}Pr_xRuO₃ increases the structural distortion, specifically, the Ru-O-Ru bond angle reduces further from 180°.³⁰ The same chemical substitution in La_{1-x}Pr_xNiO₃ leads to a metal-insulator transition at the Néel temperature in PrNiO₃.³³ As x increases in La_{1-x}Pr_xRuO₃, the inverse $\chi(T)$ lowers progressively whereas the slope does not show an obvious change in Fig.1(a). The $\mu_{\text{eff}}(T)$ of La_{1-x}Pr_xRuO₃ changes very little with x near room temperature as shown in Fig.1(b). The $\mu_{\text{eff}}(T)$ of La_{0.6}Pr_{0.4}RuO₃ can be fit to the Kotani's theory with $\lambda = 0.05$ eV over the entire range of temperature. An increased λ in the Pr-doped sample relative to that for LaRuO₃ is consistent with the bandwidth reduction reflected in the high resistivity in the Pr-doped samples in Fig.2. For PrRuO₃, there is a small amount of the impurity phase Pr₂Ru₂O₇ (see Fig.S1 in SM) which contributes to the anomaly at 160 K and an upturn below 50 K in the $\chi^{-1}(T)$ of Fig.1(b). The question here is why the magnetic susceptibility of metallic LaRuO₃ fits the model of single-ion physics. The physical picture of the anomalous metal could be uncovered through the analysis of transport properties and perhaps more intuitively from the thermal conductivity.

Fig.2(a) shows the resistivity $\rho(T)$ of La_{1-x}Pr_xRuO₃. The Pr doping in La_{1-x}Pr_xRuO₃ leads to an evolution from a metal in LaRuO₃ to an insulator in PrRuO₃; In addition to having an activated behavior, the $\rho(T)$ of PrRuO₃ increases by nearly two orders in magnitude at 2 K. Plotted on a linear scale in Fig.1(b), the $\rho(T)$ of LaRuO₃ does not behave like a normal metal. There is a clear inflection point of $\rho(T)$ near $T^* = 30$ K. Fitting the $\rho(T)$ of LaRuO₃ below 10 K to a power law $\rho(T) = \rho_0 + AT^n$ gives $n = 1.9$ which implies a Fermi liquid state (this trend persists even down to

50 mK, see Fig.S8 in SM). The curvature change of $\rho(T)$ at 30 K may resemble the coherent to incoherent state transition predicted for a Hund metal and observed in the iron pnictide superconductors.¹⁻⁴ The transition temperature T^* also coincides with the temperature where $\mu_{\text{eff}}(T)$ deviates from Kotani's theory on cooling through T^* .

Fig.3 (a) shows the thermoelectric power $S(T)$ of LaRuO_3 . The overall temperature dependence fits the description for a metal, *i.e.* $S \approx C_e/e = (\gamma/e)T$ at $T \ll \theta_D$ (Debye temperature) and the slope being reduced to $\sim(\gamma/3e)$ at high temperatures.⁴¹ Based on the diffusive formula $S = \left(\frac{\pi^2 k^2}{e E_F}\right)T$ and $E_F \approx 2eV$ from the band structure calculation in Fig.4 and Fig.S9 in the SM, a $S \approx 0.037 T$ ($\mu\text{V/K}$) is obtained. A much-enhanced slope in the $S(T) = 0.36T$ $\mu\text{V/K}$ at low temperatures indicates a strong renormalization which gives rise to a flat band at E_F due to strong correlations, which is supported by a much-enhanced γ of electronic contribution to the specific heat relative to that from the DFT calculation (discussed below). $S(T)$ is perfectly linear at low temperatures; a deviation from the linear behavior occurs above 30 K which coincides with T^* where the $\rho(T)$ goes through an inflection point. A transition from a linear $S(T)$ to a non-linear $S(T)$ appears to be relevant to the coherent state to incoherent state transition. A $S \sim 40$ $\mu\text{V/K}$ for LaRuO_3 is the largest thermoelectric power at room temperature among the single valent metallic perovskites,⁴² which could be justified by the Fermi energy at the top of the t_{2g} orbital band, see the band calculation result with DFT+U in Fig.S9. Whereas there is a crossover from a metallic phase in LaRuO_3 to an insulating phase in PrRuO_3 , as shown by a huge increase of the resistivity in Fig.2, Pr substitution in $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$ only leads to a modest increase of the thermoelectric power and no obvious change in the temperature dependence as shown in inset of Fig.S5, which is in sharp contrast to a huge increase on crossing the metal-insulator transition in the single-band system RNiO_3 .⁴⁴⁻⁴⁵ **The changes of resistivity and the thermoelectric power at 100 K on crossing the metal-insulator transition as R=La is replaced by Pr in perovskite systems RNiO_3 and RRuO_3 are shown as an inset of Fig.3(a).** The thermoelectric power in an insulator is sensitive to the charge carrier density. A drastic change of S on crossing the metal-insulator transition from LaNiO_3 to PrNiO_3 at 100 K corresponds to a gap opening at the Fermi energy. In contrast, a gradual change of S from metallic LaRuO_3 to an insulating phase of $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$ implies that the crossover from a metal to an insulator is driven by disorder. As the bandwidth narrows with increasing Pr doping in $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$, the transportation of charge carriers changes from scattering in the

metallic phase with incoherent electrons to diffusion or hopping in the insulator phase with localized electrons.

The thermal conductivity measurement provides further useful information for characterizing the incoherent state and the coherent to incoherent state transition. Fig.3(b) shows the lattice thermal conductivity κ after correcting for the electronic contribution via the Wiedmann-Franz law. The $\kappa(T)$ of LaRuO₃ at room temperature is much lower than that of typical single valent insulator perovskites like LaGaO₃⁴⁶ and metallic perovskites like LaNiO₃ and LaCuO₃.⁴³ For $T > 100$ K, the $\kappa(T)$ of LaRuO₃ behaves glassy. In a crystal, Cahill *et al*⁴⁷ have shown that disorder can significantly suppress the $\kappa(T)$; the low limit of $\kappa(T)$ in a disordered crystal can reach the level for amorphous materials. Disorder in the perovskite LaRuO₃ can be induced by impurities in the lattice or arise because of strong electron-phonon interactions, which leads to polaron physics. A sharp upturn of $\kappa(T)$ below 60 K rules out the first possibility. The upturn of $\kappa(T)$ near T^* implies that the phonon thermal conductivity is restored in the coherent state of electrons where the electron-phonon becomes relatively weak. In contrast, electrons in the incoherent state behave like large polarons which interact strongly with the lattice so as to significantly reduce the phonon mean free path to the level of interatomic distance. As a result, $\kappa(T)$ is suppressed in the incoherent phase. The high temperature limit T^* for the coherent electrons is determined by the bandwidth. T^* drops upon the Pr substituting, which explains why there is no upturn of $\kappa(T)$ in all Pr doped samples and the end member PrRuO₃. The hopping conduction leads to a further decrease of $\kappa(T)$ in all the samples of La_{1-x}Pr_xRuO₃ with $x > 0$.

The band structure of LaRuO₃ from DFT calculation in Fig.4 and Fig.S9 indicates the multiband crossing at the Fermi energy, which makes LaRuO₃ legitimate as a Hund metal. The specific heat measurement shown in Fig.S7 can be fit to the formula $C_p = \gamma T + \beta T^3$ at low temperatures with $\gamma = 41$ mJ/(K² mole). A large ratio of $\gamma/\gamma_0 \approx 16$, with $\gamma_0 = 2.6$ mJ/(K² mole) obtained from the band structure in Fig.4, provides strong evidence for the importance of Hund's coupling in LaRuO₃ as predicted theoretically by Haule and Kotliar.¹

The Bloch function for describing electrons in a solid consists of plane wave and atomic wavefunction contributions. The concept of quasiparticles addresses the many-body correlations, but the quasiparticle lifetime is subject to the on-site correlation energy U . In the presence of

strong Hund's coupling, the quasiparticle lifetime is severely modulated in a multiple-orbital system depending on the band filling number n . In a cubic perovskite oxide model system with a partially filled, low spin π^* band, Medici *et al*³ have provided the numerical solution of the Kanamori Hamiltonian. For $n=1,2, 3,4$ the effect of Hund's coupling is to increase the threshold U_c for the Mott transition, whereas U_c is much reduced for $n=3$. A characteristic temperature T^* in the phase diagram marks the temperature above which the quasiparticle lifetime becomes sufficiently short and the coherent state is lost. In the incoherent state, while metallicity remains, the probability to find charge carriers within the atomic wavefunctions is significantly high. As a result, the screening on magnetic moments becomes weak and a Curie-Weiss law is expected in the paramagnetic susceptibility.¹ The spin freezing for the incoherent electrons has been derived in a calculation by Wermer *et al.*² Specifically, Haule and Kotliar¹ have shown the influence of Hund's rule coupling in $\chi(T)$ and $\rho(T)$ for iron oxypnictides. For $J_H=0.4$ eV, the transition from the incoherent state to the coherent state is represented by the drop of resistivity. A Curie law behavior is predicted for the magnetic susceptibility in the incoherent phase with $J_H > 0.4$ eV. These authors have also shown that the electronic contribution to the specific heat γ is greatly enhanced as J_H increases.

LaRuO₃ has the low spin d^5 electron configuration in a nearly cubic crystal field. Its physical properties are all consistent with the theoretical predictions for a Hund metal. Whereas a Curie-Weiss law behavior or highly localized moment in the incoherent state has been predicted for a Hund metal, experimental verification has not been made in any systems reported in the literature. LaRuO₃ is the only metal showing the paramagnetic susceptibility fitting a model of single-ion physics. In addition, disorder in the incoherent electrons transfer into the phonon subspace via strong electron-phonon interactions in this narrow-band system, which destroys the phonon thermal conductivity. As shown in the theoretical calculation,³ U_c for a d^5 system is large. An increase of U by the Pr substituted La_{1-x}Pr_xRuO₃ is insufficient to trigger a Mott transition. Instead, the gradual crossover from a metallic $\rho(T)$ in LaRuO₃ to an activated $\rho(T)$ in PrRuO₃ is more likely due to Anderson localization by disorder. This argument is supported by a gradual change of the thermoelectric power in the crossover from metallic LaRuO₃ to an insulator PrRuO₃, which is in sharp contrast to what is found in the single-band system of RNiO₃ which exhibits a Mott transition as the bandwidth is reduced.

In conclusion, the strong intraatomic interaction restores the Hund's coupling J_H in metallic LaRuO_3 . A strong J_H reduces the quasiparticle lifetime, which is manifested by a transition from a coherent to an incoherent electronic phase from the measurement of resistivity. Moreover, the effect of J_H is to create localized spins in the incoherent phase. In LaRuO_3 , the paramagnetic susceptibility in the incoherent phase can be described by a model of single-ion physics. Since the threshold U_c for the Mott transition is significantly enlarged for the low spin system with $n=5$ due to J_H , the bandwidth reduction in $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$ series does not lead to a Mott transition, but an evolution from the metallic phase of incoherent electrons to an insulating phase by disorder. Electrons in the incoherent phase of LaRuO_3 disturb the phonon structure due to a strong electron-phonon coupling, which leads to a glassy thermal conductivity. The phonon thermal conductivity is restored in the coherent phase below T^* . All these observations make LaRuO_3 a representative Hund metal.

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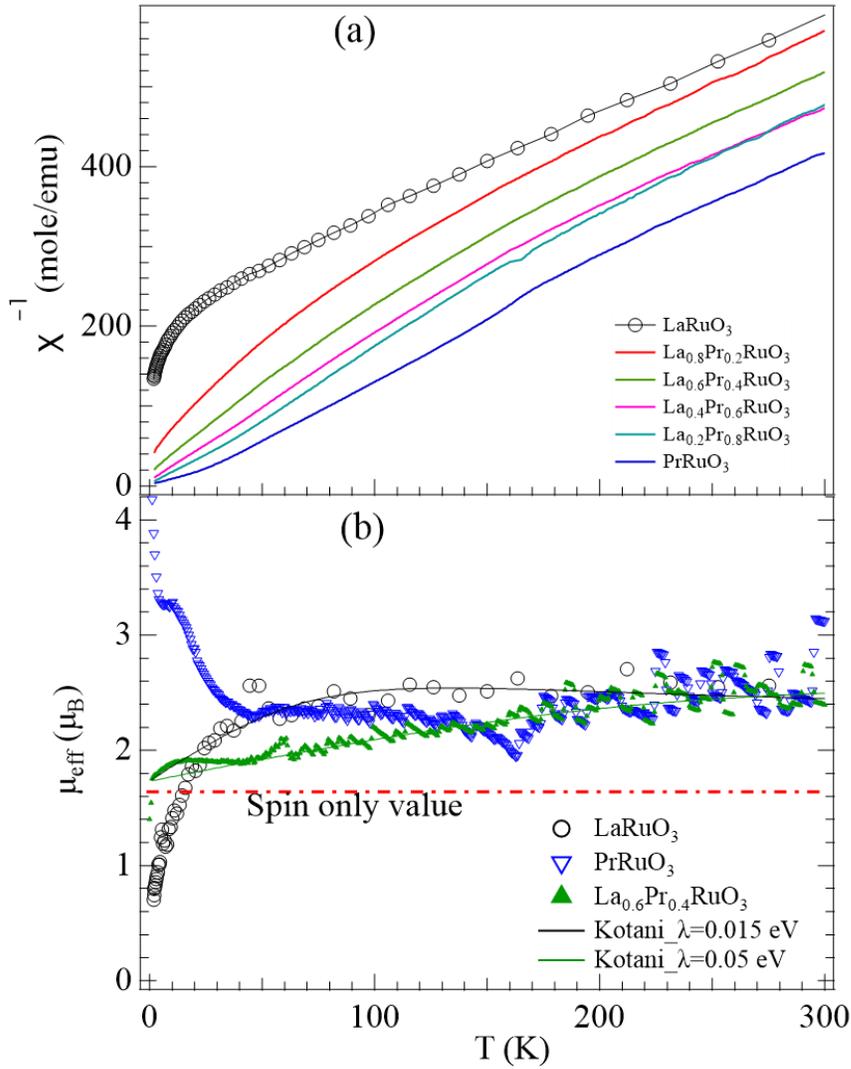


Fig.1 (a) Temperature dependence of (a) inverse magnetic susceptibility of $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$ and (b) $\mu_{\text{eff}} = \sqrt{8C}$ derived from (a) in the plot of $\chi = C/T$. Solid lines in (b) from Kotani's theory with different value of spin-orbit coupling. The dash-point line in the plot is the spin-only value of μ_{eff} in a Curie law for a system with $n=1$ or 5 .

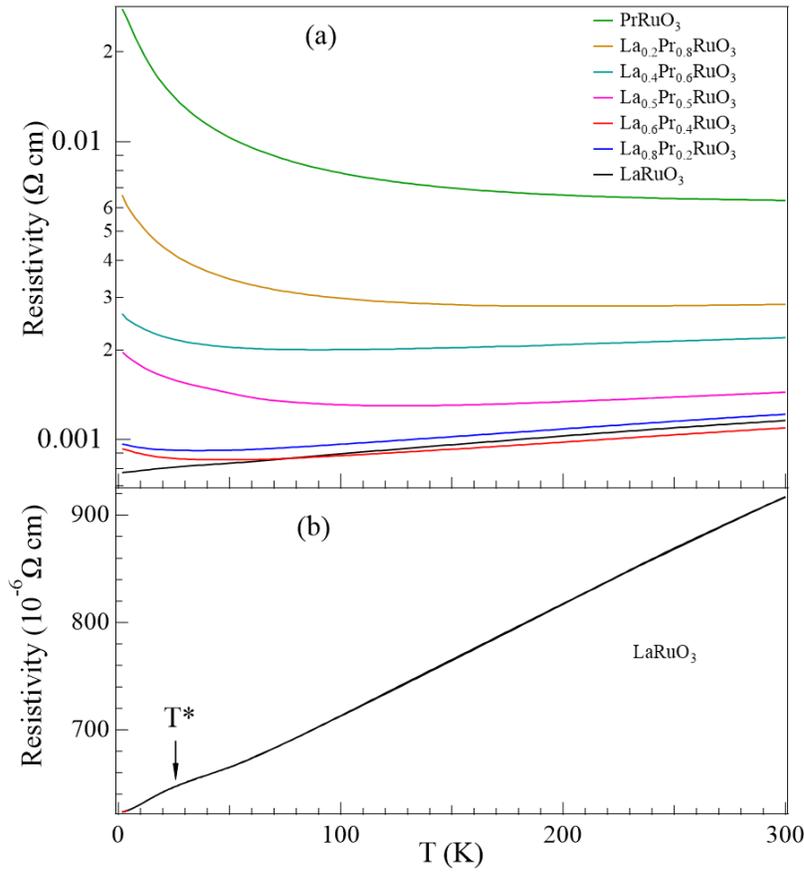


Fig.2 Temperature dependence of the resistivity in $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$. A slightly higher resistivity of LaRuO_3 than that of $\text{La}_{0.8}\text{Pr}_{0.2}\text{RuO}_3$ at room temperature may be caused by the geometry uncertainty of the samples and the distance between the voltage leads in the four-probe measurement. The resistivity drop of LaRuO_3 on cooling through T^* in the linear scale plot shown in (b) resembles the feature expected for the incoherent to coherent state transition as predicted by the theory.

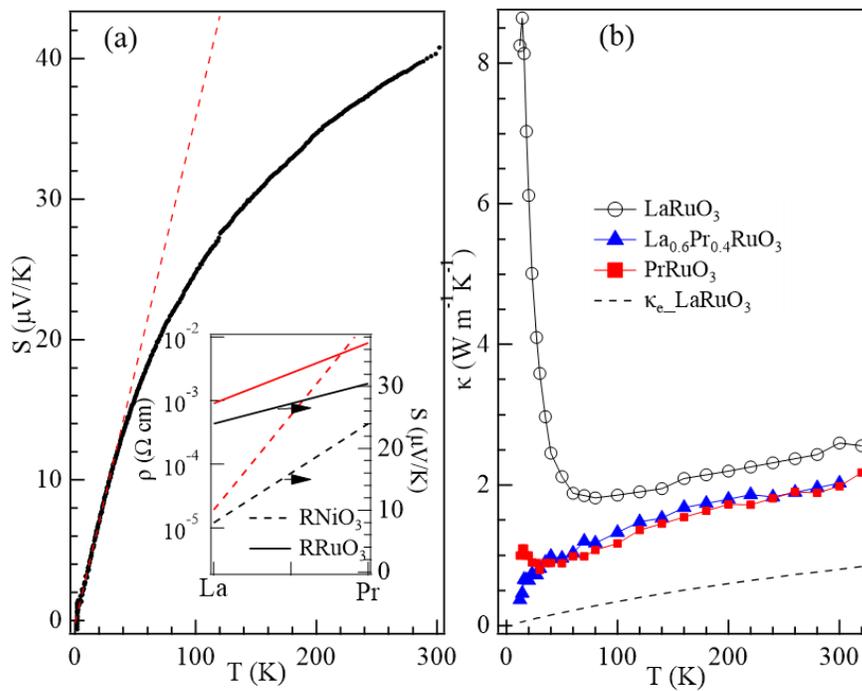


Fig.3 Temperature dependence of (a) the thermoelectric power for $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$; the inset shows the changes of resistivity and thermoelectric power at 100 K from $R = \text{La}$ to Pr in RRuO_3 and RNiO_3 and (b) the thermal conductivity of $\text{La}_{1-x}\text{Pr}_x\text{RuO}_3$.

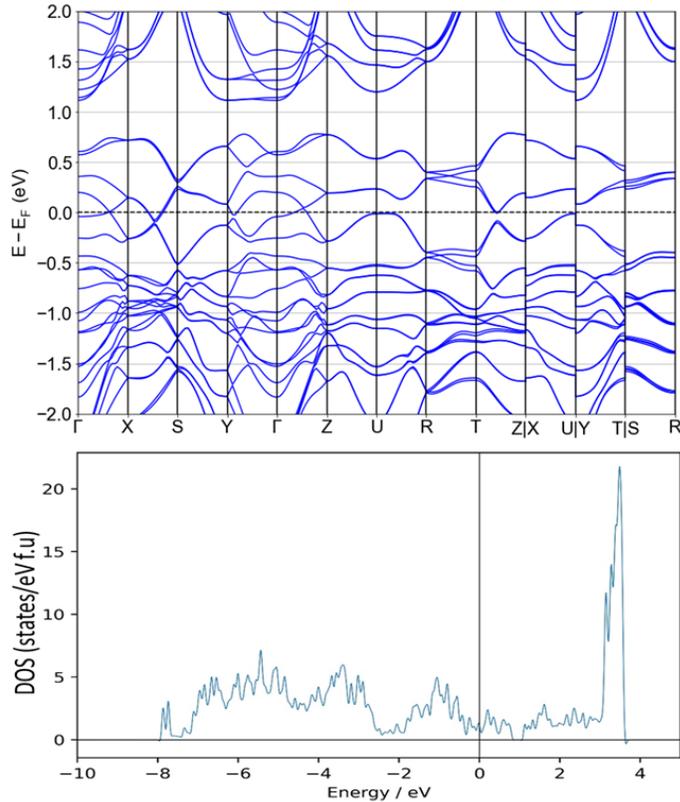


Fig.4 The band structure (upper panel) and the density of states of LaRuO_3 (lower panel) from the DFT calculation. The band structure shown here has been obtained by adding U and spin-orbit coupling (SOC) in the calculation. The calculation without SOC gives rise to an insulator of LaRuO_3 , see Fig.S9 for the result and the detailed information about the calculation.

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