

# Pressure induced ferromagnetic metal for a Mott insulator $\text{Ca}_2\text{RuO}_4$

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## Abstract

We show that application of very modest pressures to pure  $\text{Ca}_2\text{RuO}_4$  transforms it from an antiferromagnetic Mott insulator to a quasi-2D metal with a ferromagnetic ground state. The smallness of the moment compared with the saturated moment of the localized  $\text{Ru}^{4+}$  ion reflects the itinerant character of the magnetism.

*Key words:* Mott transition,  $\text{Ca}_2\text{RuO}_4$ , pressure effect, AFM-FM transition

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As well known, the quasi-two-dimensional (Q2D) Mott transition systems, especially Ru214 systems, show the rich variety of exotic magnetic and electronic properties: for instance, insulator-metal (IM) transition, spin-orbit coupling, ferromagnetism, antiferromagnetism (AF) and unconventional SC.  $\text{Sr}_2\text{RuO}_4$  (SRO) exhibits SC below 1.5K [1], and it is widely believed that the Cooper pairs form in a triplet state [2]. Moreover, many normal-state properties of SRO are interpreted in terms of a Q2D metal in the vicinity of a Mott transition [3,4]. In contrast,  $\text{Ca}_2\text{RuO}_4$  (CRO), which has a larger  $U/W$  than SRO, is a Mott insulator below 357K with antiferromagnetic (AF) ordering below  $T_N = 113\text{K}$  as indicated by a number of measurements [5]. Previous work has paid much attention to  $\text{Ca}_{2-x}\text{Sr}_x\text{RuO}_4$  (CSRO) as a unique system to link a Mott insulator to a Fermi Liquid with SC through tuning  $U/W$  [6]. To explore the underlying ground state and possibly a new SC phase in this system, pressure is generally to be preferred to chemical doping because pressure does not introduce disorder though a chemical doping is an effective and convenient method to control  $U/W$ . Here, we report resistivity and susceptibility measurements in CRO under hydrostatic  $P$ .

We measured the in-plane and out-of-plane resistivities  $\rho_{ab}$ ,  $\rho_c$  using single crystal CRO. The pressure variations at 300K are plotted in Fig. 1. With pressurising up to 0.5 GPa, both  $\rho_{ab}$  and  $\rho_c$  decrease monotonically. At 0.5 GPa  $\rho_{ab}$  and  $\rho_c$  drop discontinuously by approximately four and two orders of magnitude, respectively. Above the discontinuous drop, however,  $\rho_c$  turns to increase whereas  $\rho_{ab}$  decreases monotonically. Moreover, the anisotropic ratio  $\rho_c/\rho_{ab} \sim 2$  in the insulator phase jumps to about 100 in the metallic phase. The discontinuity indicates that the insulator-metal (IM) transition at 0.5 GPa is of first order.

The IM transition is interpreted in terms of analogy with the phase transition in pure CRO at 360K. It is reported that in pure CRO above 360K the metallic phase is L-Pbca structure, with smaller volume and less distortions than the  $T < 360\text{K}$  insulating S-Pbca phase [8]. Application of  $P$  will stabilise the smaller volume phase, so we expect that at the IM transition CRO also changes from S-Pbca to L-Pbca.

The inset shows  $\rho_{ab}$  and  $\rho_c$  as a function of temperature. At  $P < 0.5\text{ GPa}$ , both  $\rho_{ab}$  and  $\rho_c$  show an activation-type increase. In contrast, the pressurised CRO (2GPa) shows a Q2D metallic nature above  $\sim 20\text{K}$ . That is,  $\rho_{ab}(T)$  shows metallic ( $d\rho_{ab}/dT > 0$ ) conduction whereas  $\rho_c$  exhibits a weak increase indicating a nonmetallic conduction. The anisotropy ratio

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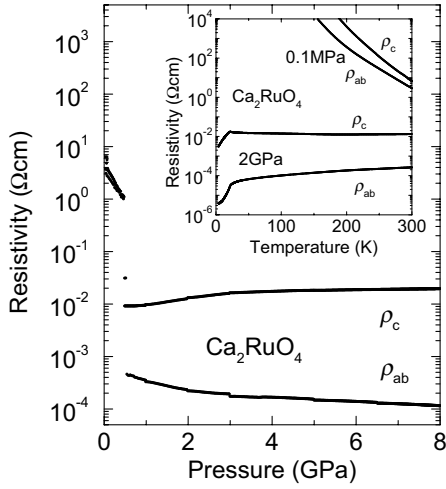


Fig. 1.  $\rho_{ab}$  and  $\rho_c$  measured at 300K with increasing pressure up to 8GPa. The discontinuous drop at 0.5GPa indicates an Mott transition. Inset shows temperature dependence of  $\rho_{ab}$  and  $\rho_c$  measured under 0.1MPa and 2GPa.

reaches up to about 1000 at low  $T$ . Thus, the Q2D nature of the metallic state is comparable with HTC or SRO.

Here we can see another prominent feature that both  $\rho_{ab}(T)$  and  $\rho_c(T)$  curves at 2GPa shows remarkable reduction below  $T^* \sim 20$ K.  $\rho_c(T)$  turns from nonmetallic increase to a rapid decrease indicating some transition. At the  $T^*$  corresponding to the kink in the  $\rho_c(T)$  curves,  $\rho_{ab}(T)$  also shows a rapid reduction towards a residual resistivity as low as  $3 \mu\Omega \text{ cm}$ . Generally speaking, a reduction of resistivity observed in a magnetically ordered state is interpreted in terms of a suppression of the magnetic scattering. To clarify what is happening in  $P$ -induced metallic CRO below  $T^*$ , the magnetization measurements were performed under  $P$  using a piston-cylinder clamp cell with a commercial SQUID magnetometer (Quantum Design, model MPMS).

Figure 2 shows magnetization-field ( $M$ - $H$ ) curves measured at 2 K with  $H_{\perp c}$  under ambient pressure (AF insulating phase) and 0.7GPa (metallic phase). A hysteresis indicating a FM ordering can be seen in the curve at 0.7GPa although no hysteresis was observed at ambient pressure. The remnant magnetisation is estimated to be about  $0.09 \mu_B$ . Nevertheless, the itinerant character of the magnetism is indicated by the quite small moment than the saturated moment  $2\mu_B$  of the localized  $\text{Ru}^{4+}$  ion.

The  $M$ - $H$  curves were also measured at several fixed temperatures under  $P = 0.7$  GPa. The hysteresis loop was observed below  $\sim 12$  K, which is comparable to  $T^*$  near 1 GPa. Thus, we can understand that the remarkable drop in  $\rho(T)$  at  $T^*$  is most probably due to

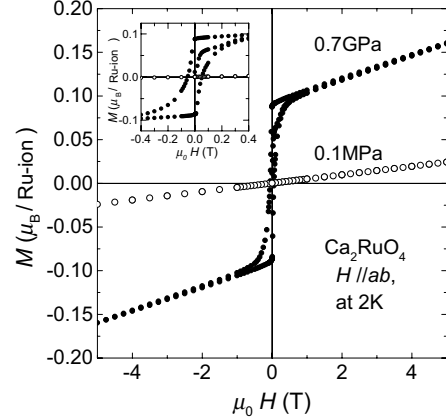


Fig. 2. Magnetisation curves for 0.1MPa and 0.7GPa, measured at 2K. Hysteresis indicating FM ordering is observed at 0.7GPa, where the system is metallic in the corresponding resistivity measurement.

reduced magnetic scattering at a FM transition.

In conclusion, it is indeed tempting to see that the pressurised CRO exhibits a wide variety of interesting phenomena, especially a transition from AF insulator to a Q2D metal with a FM ground state. From analogy with CSRO, we infer that the complicated electronic and magnetic properties might be strongly coupled to structural changes. The occurrence of actual FM ordering is unique and does not found in the doped system where disorder is introduced. Intrinsic phase-diagram thus appear only under  $P$ [9]. Much attention will have been paid to the comparison of the properties of the itinerant-FM in the Q2D-system with theoretical predictions [10]. Moreover, strong interest will be paid for the measurements at higher pressures as a crucial test of the connection between the triplet SC of SRO, and the itinerant FM of CRO.

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