Localization Crossover Near Metal-Insulator Transition in Two-Dimension Limit of \( \text{CaCu}_3\text{Ru}_4\text{O}_{12} \)

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Artificial confinement of electrons by tailoring the layer thickness has turned out to be a powerful tool to harness control over competing phases in complex oxides. We investigate the effect of dimensionality and strain on the electronic band structure and transport properties of \( d \)-electron based heavy-fermion metal \( \text{CaCu}_3\text{Ru}_4\text{O}_{12} \). Transport behavior evolves from metallic to localized regime upon reducing thickness and a metal insulator transition is observed below 3 nm for which sheet resistance crosses \( h/e^2 \sim 25 \) KΩ, the quantum resistance in 2D. A strong interplay between inelastic and spin-orbit scattering lengths close to metal insulator transition is observed which results in a weak antilocalization to weak localization crossover in magnetococonductance upon reducing film thickness. The cause of metal-insulator transition and magnetoconductance crossover are explained using band structure calculation and 2D magnetotransport theory.

Reducing the dimensionality of a system often engenders electromagnetic properties sharply different from their bulk counterpart. It mainly arises due to enhanced quantum effects and increased correlations due to reduction in available phase space and screening. The interplay between electron band width \( (W) \) and onsite Coulomb energy \( (U) \) in correlated electron system plays a crucial role in addressing the metal-insulator transition (MIT) [1]. The control of \( W/U \) ratio in correlated electron system sensitively depends on the dimensionality of the electron system, which can precisely be tailored with the advances in thin-film growth technology. The study on dimensionality controlled MIT has been realised in 3d transition metal oxide based thin films by controlling the layer thickness [2,5] and recently it has been extended to spin-orbit( SO) coupled \( 5d \) iridates where novel topological phases are revealed [6,7]. The possibility to manipulate the electronic state of correlated electrons in artificial crystal structures by exploiting their layer thickness and epitaxial strain (without resorting to any chemical substitution that might induce unintentional disorder) is seen as a viable route to obtain more insight into these materials and renders a perfect platform to search for unforeseeable complex phenomena [2,11].

In this letter, we focus on the study of the dimensionality effect on A site ordered perovskite \( \text{CaCu}_3\text{Ru}_4\text{O}_{12} \) (CCRO) which is intriguingly debated as a rare class of \( d \)-electron based heavy-fermion system along with a signature for broad \( \chi(T) \) hump around 150-200 K[12]. Traditionally, \( f \)-electron-based heavy-fermion states in metallic rare earth compounds are formed \( \text{via} \) hybridization between localized \( f \)-electrons and itinerant band electrons. On a similar footing it was proposed for CCRO that electrons in the Ru-4\textit{d} (Ru\textit{t}\textit{d} : 4\textit{d}⁰) derived conduction band hybridises with the localised Cu (Cu\textit{t}\textit{d} : 3\textit{d}⁰) magnetic moments, resulting in a Kondo lattices [13]. However, the Kondo scenario was later dismissed as no localized magnetic moment was observed in neutron scattering experiment up to 700 K [14], but the origin of \( \chi(T) \) broad maximum has remained elusive.

Besides rich and anomalous magnetic properties, bulk CCRO is considered to be a correlated metal with excellent metallic conductivity [15] and exhibits non Fermi Liquid behaviour below 2 K [14]. Though there have been studies on bulk poly-crystalline CCRO, the investigation on single-crystalline bulk/thin film is rare. In this study, we fabricate single crystalline epitaxial CCRO thin films by varying the layer thickness over a wide range and address the effect of dimensionality on its electronic band structure and magnetococonductivity. We observe that upon reduction of CCRO thickness, transport behavior evolves from metallic to localized regime and a thickness driven metal insulator transition (MIT) is observed below 3 nm for which the room temperature sheet resistance crosses \( h/e^2 \sim 25 \) kΩ, the quantum resistance in 2D [2,6]. The band structure calculations reveal that reduced thickness is accompanied with certain structural distortion which augments the strong correlation effect to localize the states and in turn to induce MIT. More importantly, from magnetococonductance we observe a strong inter-play among elastic, inelastic (Thouless) and spin-orbit scattering lengths \( (l_e, l_{Th}, l_{So}) \) close to MIT that gives rise to weak antilocalization (WAL) - weak localization (WL) crossover upon reducing thickness. Using general 2D magnetotransport theory, we present plausible physical explanation for WAL-WL crossover as a function of thickness.

A series of high-quality single-crystalline epitaxial CCRO films with varying thickness \( (t) \) were grown on the LaAlO\textsubscript{3} (001) substrates \( (a = 3.79 \) Å) using pulsed
laser deposition. Details of deposition method are described in Supplemental Material (SM) [16]. Bulk CCRO exhibits cubic symmetry (Im-3 space group (No: 204)) with a lattice parameter of 7.43 Å [14] which is close to twice the lattice parameter of LaAlO3 (LAO)(Fig.1(a)). Thus, one can expect an in-plane tensile strain of +1.97 % for CCRO films with a cube-on-cube epitaxy on LAO. Fig.1(b) exhibits the diffraction pattern around (002) reflection of LAO for various thickness of CCRO films. A full scan of XRD pattern is provided in SM. It is noted from Fig.1(a) that the position of CCRO (004) reflection shifts systematically away from substrate peak and gets broadened upon reducing film thickness. The variation of c-axis (out-of-plane) lattice parameter as a function of thickness indicates that c relaxes close to the bulk value for films with t ≥ 12 nm, where as it shrinks progressively towards lower thickness regime signalling a compressive out-of-plane strain (Fig.1(c)). The close match of c-axis lattice parameter with bulk value for thicker films indicate about the optimal oxygen stoichiometry. To verify the in-plane epitaxial relationship, the ϕ scans were performed (Fig.1(d)). Four equally spaced distinct peaks with a relative separation of 90° (four-fold symmetry) were observed suggesting the cube-on-cube epitaxial growth of CCRO layer on LAO i.e:[100]LAO||100]CCRO. From reciprocal space mapping(see SM), the in-plane lattice parameter for 60 nm film is estimated to be 7.44 ± 0.05 Å, a value that closely matches to the polycrystalline bulk CCRO value [14], indicating the film to be relaxed. However, the 6 nm film is tensile-strained exhibiting an increased in-plane lattice parameter of 7.64 ± 0.05 Å. Altogether, our structural characterization implies an epitaxial strained growth of CCRO lattice near the interface up to 6 nm, and a relaxed growth for thickness above 12 nm on LAO. From XPS study (see SM), formal valence of Cu and Ru are found to be +2 and +4 respectively which is consistent with literature [17].

We turn our discussion now to the electrical transport properties of CCRO films with thicknesses (t) ranging from 1.5 to 60 nm. Fig.2 shows the variation of sheet resistance (R_S) and resistivity (ρ) as a function of temperature. For films with t ≥ 3 nm, R_s values are found to be below 25 kΩ (h/e^2 ≈ 25 kΩ) across the whole temperature range (2–300 K). However, the value of R_s for the thinnest film with t = 1.5 nm crosses 25 kΩ indicating a transition to insulating state. Based on Ioffe and Regel criterion MIT is expected in the limit of k_Fl_c ≈ 1 where k_F, l_c are Fermi wave vector and mean free path respectively [18, 19] and an approximate value for the Mott-Ioffe-Regel limit in two dimensions is h/e^2 [20]. The systematic increase in R_s and its rise above Ioffe-Regel limit indicates the occurrence of localization effect as the thickness of CCRO film is reduced. Transport in 1.5 nm film follows Arrhenius type behavior σ ∝ exp(-E_o/2kT) and yields an activa-
tion energy gap $E_g$ of 32 meV, which is obtained by fitting the data in the temperature range 300-150 K. On the contrary for the case of 3 nm film, which exhibits a negative temperature coefficient ($dp/dT < 0$) and is on the verge MIT, the transport behavior can be described by a variable range hopping (VRH) type of conduction. VRH conduction scenario involves hopping of electrons between the localized electronic states within narrow bands close to the Fermi energy and conductivity $\sigma$ is given by $\sigma = C \exp \left[-\left(T_0/T\right)^{\alpha}\right]$ [21, 22], where $T_0$ depends on the density of localized states and spread of wave functions. VRH conductivity can either be of either of Mott or Efros-Shklovskii (ES) type and for a two dimensional case they are characterized by the exponent $\alpha = 1/3$ and $\alpha = 1/2$ respectively [21, 22]. In our case for 3 nm CCRO film, the fit to the conductivity data in the temperature range 20 K $\leq T \leq$ 225 K yields $\alpha = 0.502$ (ES type) indicating the existence of Coulomb charge gap. At higher thickness side, the temperature dependent metallic resistivity is well described by $\rho = \rho_0 + AT^2$ relation in the temperature range of 2-50 K ($\rho_0$ is residual resistivity and the coefficient $A$ quantifies the electron–electron interactions), a characteristic of Fermi liquid behavior in which $e-e$ scattering cross section varies quadratically with temperature. Fig.2 (d) shows $\rho$ vs $T$ plots that fits reasonably well to $T^2$ and is in agreement to what is observed by Krimmel et al. in bulk CCRO [13]. The values of the fitting parameters are found to be $\rho_0 = 0.359 \times 10^{-3}$ $\Omega$cm, $A = 3.1 \times 10^{-9}$ $\Omega$cm K$^{-2}$ and $\rho_0 = 0.297 \times 10^{-3}$ $\Omega$cm and $A = 2.4 \times 10^{-9}$ $\Omega$cmK$^{-2}$ for 23 and 60 nm thick CCRO films respectively. It may be noted that electron-magnon scattering cross section also follows a $T^2$ dependence, but in this case latter effects contribution is not considered since CCRO is a very weak paramagnet.

To examine the changes in electron scattering process upon approaching MIT induced by thickness variation, we performed magnetotransport measurements. Constructive (destructive) interference among the scattered electrons which propagate in identical time-reversed closed trajectories give rise to WL (WAL) effect and these quantum effects can be captured by measuring conductivity in presence of magnetic field. Fig.3(a) shows the out of plane magnetoconductance (MC) $\Delta\sigma$ in units of $e^2/\pi\hbar^2$ measured at 2 K for film thicknesses ranging from 60 to 3 nm. A negative MC is observed for films with large thickness ( $t \geq 6$ nm). However, a crossover from negative to positive MC occurs as we approach the MIT. We attribute the positive (negative) magnetoconductance to WL (WAL) effect. To shed light on the observed magnetoconductance crossover, we fit the magnetoconductance curves with Hikami-Larkin-Nagaoka (HLN) equation [23] in 2D limit to extract the characteristic diffusion lengths. For the convenience of experimental data fitting, we can express modified form of HLN equation (Eq.(1)) in 2D limit in terms of elastic, inelastic and spin-orbit scattering lengths which are denoted as $l_e$, $L_{Th}$, $l_{so}$ respectively.

\begin{equation}
\Delta\sigma = -\frac{e^2}{2\pi\hbar^2}\left[\psi(1/2 + B_e/B) + \log(B/B_e)\right] - \frac{1}{2}\left(\psi(1/2 + B_{Th}/B) + \log(B/B_{Th})\right) + \frac{3}{2}\left(\psi(1/2 + \frac{B_{Th} + 4B_{so}}{B}) + \log(\frac{B}{B_{Th} + 4B_{so}})\right)
\end{equation}

where $B$ is external magnetic field and $B_e, B_{Th}, B_{so}$ are related with $l_e, L_{Th}, l_{so}$ respectively as following relations $B_i = \hbar/4e l_i^2$.

We define 2D limit for film thickness (t) which is less than $L_{Th}$ ($L_{Th} = (D\tau_0)^{1/2}$); where $D$ is Diffusion coefficient and $D \propto v_F^2$ ($v_F$ is Fermi velocity) and $\tau_0$ is inelastic scattering time [24]. Fig.3(c and d) show the fitted curves as per Eq.(1) and the extracted lengths from the fitting are shown in Fig.3(d). The extracted lengths show a crossover from $L_{Th} \geq l_{so}$ for the films with $t < 6$ nm to $L_{Th} \leq l_{so}$ for $t > 6$ nm. Below we discuss case by case study for the observed magnetoconductance crossover with thickness variation.

It is evident from Fig.3(b) that 3 nm thick film shows WL effect (i.e. $\Delta\sigma(B)$ monotonically increases with increment of magnetic field up to 8 Tesla). Since $L_{Th} < l_{so}$ for 3 nm case, the spin-orbit related scattering effect becomes redundant (spin-orbit interaction strength $\propto 1/l_{so}^2$) for the phase space in which quantum coherence is maintained, electron encountering SO scattering is very weak. In other words, small $L_{Th}$ allows Cooperon loop in very short length scale where SO interaction is not able to rotate the spin and thus results in WL due to constructive interference. However, in 6 nm thick film with...
$L_{TH} \sim l_{so}$ results in a crossover from WL to WAL with variation of external magnetic field as shown in Fig.3(b) ($\Delta\sigma(B)$ increases with magnetic field up to $\sim 2.5$ T and then it starts to decrease). In WL regime, magnetic flux induces additional phase difference between two electrons moving in time-reversed identical closed trajectory and this additional phase difference breaks constructive interference and as a result conductivity increase with the application of magnetic field. For a particular magnetic field $B_{max}$, additional phase difference breaks completely the constructive interference for which $\Delta\sigma(B_{max})$ attains a maximum and a further increment of $B$, $\Delta\sigma(B)$ starts to decrease revealing the signature for WAL. This may be plausible explanation for the observed behaviour of $\Delta\sigma(B)$ in 6 nm film which shows monotonically increasing trend with $B$ up to $(B_{max}) \sim 2.5$ Tesla and then followed by a decreasing trend with further increase in $B$. For films with $t \geq 12$ nm, $L_{TH}$ tends to increase and $l_{so}$ gradually decreases. Therefore electrons are able to move relatively more distance by maintaining phase coherence and simultaneously due to smaller value of $l_{so}$, electrons encounter spin-orbit elastic scattering within a shorter distance. Thus WAL becomes prominent with increasing thickness and is a cumulative effect of above two factors.

To gain more insight into the MIT with reduced thickness of the CCRO film, we carried out DFT+U calculations using the pseudopotential method [25] on bulk, 4, and 2 u.c. (corresponding to 1.5 nm) thick CCRO films and the relevant results are shown in Fig. 4. The computational details are provided in the SM. The densities of states (DOS) shown on the left panel (for $U = 6$ eV) indicates the formation of a pseudogap in the vicinity of the Fermi level ($E_F$). For 2 u.c the number of states ($n$) at $E_F$ per eV per formula unit decreases rapidly as we move from bulk ($n \sim 5$; see inset) [26] to 4 u.c. film ($n \sim 1.5$) to 2 u.c film ($n \sim 0.9$). Concurrently, the conductivity decreases substantially with reduced thickness which agrees well with the experimental inferences. To examine the nature of gap formation, on the right panel we have shown the evolution of the band structure for the 2 u.c. film as a function $U$. For $U = 0$ eV, the system is metallic as in the bulk. With increasing $U$, the correlation effect increases to deplete the states at $E_F$ through Mott mechanism. The structural relaxation calculations, discussed in the SM, show that with reduced thickness, there is an increase in the lattice distortion through the RuO$_6$ octahedra which introduces localization by weakening the covalent interaction. The localization strengthen the on-site repulsion, here incorporated through $U$. As a consequence, lower and upper Hubbard bands are formed out of Ru-$d$ states (see Fig.4, bottom left). In the case of 2 u.c. film, these Hubbard bands (LHB and UHB) are separated by gap.

In conclusion, we have demonstrated that epitaxial single crystalline thin films of heavy-fermion correlated metal CCRO grown on LAO can be driven to an insulating state upon reducing its thickness. Further, we find that magnetoconductance shows a crossover from WAL to WL behaviour on approaching metal insulator transition. From extensive analysis of magnetotransport data, we realize that a subtle interplay between spin-orbit elastic scattering and inelastic scattering of electrons close to metal insulator transition renders such crossover. Spin-orbit interaction scattering process plays an important role in understanding WAL-WL transition. Apparently spin-orbit interaction scattering process seems to be an intrinsic property of material, but we have shown here that spin-orbit interaction scattering length ($l_{so}$) changes systematically upon CCRO film thickness variation and this holds the major-key for magnetoconductance crossover near MIT. Band structure analysis on CCRO films reveals that the reduction in the thickness of the film distorts the RuO$_6$ complexes which weakens the covalent interaction leading to localization. The latter through strong correlation effect, following Mott mechanism, depletes the states at Fermi level leading to the experimental observation of MIT.

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Supplemental material for: Localization Crossover Near Metal-Insulator Transition in Two-Dimension Limit of CaCu$_3$Ru$_4$O$_{12}$

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I. CCRO PLD TARGET PREPARATION AND THIN FILM GROWTH

CaCu$_3$Ru$_4$O$_{12}$(CCRO) thin films were grown on single-crystalline (001) oriented LaAlO$_3$ (LAO) substrates using Pulsed laser deposition (PLD) with a KrF excimer laser ($\lambda = 248$ nm). The polycrystalline CCRO PLD-target material was prepared by stoichiometrically mixing CaCO$_3$, CuO, RuO$_2$ and heating the mixture for 26 hours at 1050 °C temperature in open air and under ambient pressure [1,2]. Single phase was obtained after many time heating and grinding with respect to the above heat treatment process. During thin film growth, the substrate temperature and the O$_2$ partial pressure was maintained at 650 °C and $5 \times 10^{-3}$ mbar respectively and the deposition was carried out with a laser fluence 2 J/cm$^2$. Before deposition the target was pre-ablated for 2 minutes to remove any possible surface contamination. After deposition, the samples were cooled down slowly to room temperature. These deposition conditions have been optimized to result in the growth of the single phase CCRO films as shown in Fig.S1. The thicknesses of the films were calibrated with the number of laser pulses using cross-sectional SEM.

II. X-RAY DIFFRACTION

Fig.S1(a) shows a wide scan $\theta$ − 2$\theta$ X-ray diffraction pattern for a CCRO film (23 nm) grown on a (001)-oriented LAO. The XRD pattern revealed only noticeable (00l) CCRO peaks without any evidence of other orientations and impurities indicating highly oriented growth of CCRO layer on LAO [1,2]. We have also carried out reciprocal space mapping of CCRO thin films to estimate the variation of in-plane lattice parameter as shown in Fig.S1(b). The in-plane lattice parameter for CCRO 60 nm thin film is found to be 7.44 ± 0.05 Å, which is completely relaxed and close to its single crystal bulk counterpart. Interestingly, the 6 nm film shows the lattice parameter of 7.64 ± 0.05 Å indicating an in-plane tensile strain at lower thickness regime.

III. X-RAY PHOTOELECTRON SPECTROSCOPY

X-ray photoelectron spectroscopy (XPS) studies were carried out using a commercial AXIS-Ultra DLD Shimadzu system with dual anode X-ray source (Mg and Al with 15kV, 30mA), Al-K$\alpha$ monochromator and a base pressure < 5 × 10$^{-10}$ mbar. Fig.S2(a) shows XPS result for the Cu 2p$_{3/2}$ core level. Distinct satellite structure (2p$^5$3d$^9$) corresponding to Cu 2p$_{3/2}$ peak indicates that copper ions have a formal valence of +2 as in the case of CuO [3]. Fig.S2(b) shows high resolution XPS plot of Ru 3d core level. It is apparent from the fitting that two peaks are present in Ru 3d spectra. This type of feature is mostly observed in RuO$_2$ indicating itinerant character of Ru 4d electrons and +4 oxidation state of Ru [4].
IV. DIFFERENT FORM OF HLN EQUATION

General expression for magneto-conductance (MC) (2D) was derived by Hikami, Larkin, Nagaoka which is known as HLN equation. The conductivity in presence of magnetic field (perpendicular system’s plane) given by \[5\]

\[
\sigma(B) = \sigma_0 - \frac{e^2}{2\pi^2\hbar} \left( \psi\left(\frac{1}{2} + \frac{1}{\tau_0}\right) - \psi\left(\frac{1}{2} + \frac{1}{\tau_1}\right) + \frac{1}{2} \psi\left(\frac{1}{2} + \frac{1}{\tau_2}\right) - \frac{1}{2} \psi\left(\frac{1}{2} + \frac{1}{\tau_3}\right) \right)
\]

(1)

where \(a = 4D_eH/\hbar c\),
\[
\frac{1}{\tau_0} = \frac{1}{\tau_z} + \frac{1}{\tau_x} + \frac{1}{\tau_y}/i\omega,
\]
\[
\frac{1}{\tau_1} = \frac{1}{\tau_z} + \frac{1}{\tau_x} + \frac{1}{\tau_y}/i\omega,
\]
\[
\frac{1}{\tau_2} = \frac{1}{\tau_z} + \frac{1}{\tau_x} + \frac{1}{\tau_y}/i\omega,
\]
\[
\frac{1}{\tau_3} = \frac{1}{\tau_z} + \frac{1}{\tau_x} + \frac{1}{\tau_y}/i\omega,
\]

where \(\psi\) is Digamma function, \(\tau_{z\alpha}, \tau_{x\alpha}, \omega\) are scattering time due to i-th component contribution of spin-orbit, localized magnetic interaction and omega related with the cutoff parameter respectively. Impurity scattering amplitude can be written in general as

\[
f_{\alpha\beta} = a_{\alpha\beta} + ib_{\alpha\beta}(\vec{n} \times \vec{n}') \sigma_{\alpha\beta} + \vec{s}_{\alpha\beta}
\]

where \(\alpha, \beta\) are spin index of scattered electrons and \(\vec{n}, \vec{n}'\) are unite vector along the wave vectors before and after scattering \(\vec{k}, \vec{k}'\) respectively. \(a, b, \vec{s}\) are non-magnetic, spin-orbit and magnetic interaction amplitudes respectively.

\[
\sigma_{\alpha\beta} = \langle \alpha | \sigma | \beta \rangle
\]

\(\sigma\) is Pauli’s matrices. Over all scattering rate (Fermi-golden rule)

\[
\frac{1}{\tau} = 2\pi\nu \sum_{\alpha,\gamma} <f_{\alpha\beta}(\vec{n}, \vec{n}')f_{\beta\gamma}(\vec{n}', \vec{n})>\nu'
\]

where \(\nu\) is density of scattering centers and \(\tau_0, \tau_{z\alpha}, \tau_{m}\) are scattering time for non-magnetic, spin-orbit, magnetic interaction respectively.

Weak localization (WL) and Weak Anti localization (WAL) occurred by interference between two identical trajectory of scattered electrons. It is known as Cooperon loop. Impurity vortex pair can be written as

\[
\Gamma = \langle f(\vec{n}, \vec{n}') \otimes f(-\vec{n}, -\vec{n}') \rangle
\]

\[
= \frac{1}{2\pi\nu\tau} - \frac{\sigma_{\alpha\beta}}{2\pi\nu\tau} + \frac{\vec{s}_{\alpha\beta}}{2\pi\nu\tau}
\]

We introduce singlet, triplet projection operators respectively

\[
S = \frac{1-\sigma_{\alpha\beta}}{4},
\]
\[
T = \frac{1+\sigma_{\alpha\beta}}{4},
\]
\[
\Gamma = \frac{S}{4\pi\nu\tau} \left( \frac{1}{\tau} - \frac{\sigma_{\alpha\beta}}{\tau} \right) + \frac{T}{4\pi\nu\tau} \left( \frac{1}{\tau} - \frac{\sigma_{\alpha\beta}}{\tau} - \frac{\vec{s}_{\alpha\beta}}{\tau} \right)
\]

Vortex \(\Gamma\) are connected by pair of Green functions

\[
\Pi(q) = \int \frac{d\rho^\nu}{(2\pi)^n} G^R(p+q) \otimes G^A(-p) \quad \text{(For n dimension) [6]}
\]

\[
= 2\pi\nu\tau \left( 1 - \tau Dq^2 \right)
\]

After taking summation over all maximally cross diagrams, the Cooperon propagator can be expressed as
\[ C(q) = \frac{\Gamma}{(1 - \Pi G)} \]
\[ = \frac{S}{2\pi\nu\tau^2} \left( Dq^2 + \frac{6}{\tau_m} \right)^{-1} + \frac{T}{2\pi\nu\tau^2} \left( Dq^2 + \frac{4}{\tau_{s0}} + \frac{2}{\tau_m} \right)^{-1} \]  
\[ C(q) = \frac{1}{(1 - \Pi G)} \]
\[ = \frac{S}{2\pi\nu\tau^2} \left( Dq^2 + \frac{6}{\tau_m} \right)^{-1} + \frac{T}{2\pi\nu\tau^2} \left( Dq^2 + \frac{4}{\tau_{s0}} + \frac{2}{\tau_m} \right)^{-1} \]

Conductivity can be obtained by taking trace on spin indexes and integration over Cooperon loop with possible transferred momentum(q) during scattering for n-dimension

\[ \Delta\sigma = -2e^2 \sum_{v} v_{s,k} (C(q)G^R(k)G^A(k)G^R(-k + q)G^A(-k + q)) \]
\[ = -\frac{2e^2\nu D\tau}{\hbar} \int \frac{(dq)^n}{(2\pi)^n} \text{Tr} \ C(q) \]  
\[ \text{Using equation no (2) and trace over singlet and triplet give -1, 3 respectively conductivity becomes} \]
\[ \Delta\sigma = -\frac{2e^2\nu D\tau}{\hbar} \int \frac{(dq)^n}{(2\pi)^n} \text{Tr} \ C(q) \]
\[ = \frac{e^2}{\pi\hbar} \int \frac{(dq)^n}{(2\pi)^n} \left[ \left( q^2 + 6l_m^{-2} \right)^{-1} - 3\left( q^2 + 4l_{s0}^{-2} + 2l_m^{-2} \right)^{-1} \right] \]

For practical situation integration limit of q has some upper and lower cutoff. Now to fix appropriate cutoff we recall simplest situation i.e (absence of spin-orbit, magnetic scattering \( l_{s0} = 0, l_m \)) integral part with cutoff for 2D

\[ \int_{(D\tau_0)^{-1/2}}^{(D\tau_0)^{-1/2}} \frac{(dq)^2}{(2\pi)^2} \frac{1}{Dq^2} = \frac{1}{2\pi D} \ln(\tau_e/\tau_0) \]  
\[ \text{where } L_{Th} = (D\tau_0)^{1/2}, \text{ elastic length } l_e = (D\tau_e)^{1/2} \]

Now we can rewrite equation (5) in a different way as given below

\[ \int_{(D\tau_0)^{-1/2}}^{(D\tau_0)^{-1/2}} \frac{(dq)^2}{(2\pi)^2} \frac{1}{Dq^2} = \int_0^{\infty} \frac{(dq)^2}{(2\pi)^2} \frac{1}{Dq^2 + (1/\tau_0)} - \int_0^{\infty} \frac{(dq)^2}{(2\pi)^2} \frac{1}{Dq^2 + (1/\tau_e)} \]  
\[ \text{Similarly equation can be written as in presence of other scattering} \]
\[ \int_{(D\tau_0)^{-1/2}}^{(D\tau_0)^{-1/2}} \frac{(dq)^2}{(2\pi)^2} \frac{1}{q^2 + l_e^{-2}} = \int_0^{\infty} \frac{(dq)^2}{(2\pi)^2} \frac{1}{q^2 + l_e^{-2} + l_{Th}^{-2}} - \int_0^{\infty} \frac{(dq)^2}{(2\pi)^2} \frac{1}{q^2 + l_e^{-2} + l_{Th}^{-2}} \]

which gives same result (for 2D only) as equation (5).

In presence of magnetic field \( q \rightarrow q + 2eA \) where \( A \) is magnetic vector potential. Dynamics is quantized due to formation of Landau levels.

\[ q_n^2 = (n + 1/2)\frac{eB}{\hbar} \]  
\[ \text{n is positive integer including zero} \]

Complete expression of equation (4) regularized with cutoff parameters ( for 2D)

\[ \Delta\sigma = \frac{e^2}{\pi\hbar} \int_0^{\infty} \frac{(dq)^2}{(2\pi)^2} \left[ \left( q^2 + 6l_m^{-2} + l_{Th}^{-2} \right)^{-1} - \left( q^2 + 4l_{s0}^{-2} + 2l_m^{-2} \right)^{-1} \right] \]
\[ + \frac{3e^2}{\pi\hbar} \int_0^{\infty} \frac{(dq)^2}{(2\pi)^2} \left[ \left( q^2 + 4l_{s0}^{-2} + 2l_m^{-2} + l_e^{-2} \right)^{-1} - \left( q^2 + 4l_{s0}^{-2} + 2l_m^{-2} + l_{Th}^{-2} \right)^{-1} \right] \]
In presence of magnetic field integration on $q$ will replace by summation and substituting $q$, $dq$ values from equation (8) we get

$$\Delta \sigma = \frac{e^2}{\pi \hbar} \sum_{n=0}^{n_{\text{max}}^2} \frac{(2\pi q_n dq_n)}{(2\pi)^2} \left( (q_n^2 + 6m_e^{-2} + l_{TB}^{-2})^{-1} - (q_n^2 + 6m_e^{-2} + l_e^{-2})^{-1} \right)$$

$$+ \frac{3e^2}{\pi \hbar} \sum_{n=0}^{n_{\text{max}}^2} \frac{(2\pi q_n dq_n)}{(2\pi)^2} \left( (q_n^2 + 4l_{as}^{-2} + 2l_m^{-2} + l_e^{-2})^{-1} - (q_n^2 + 4l_{as}^{-2} + 2l_m^{-2} + l_{TB}^{-2})^{-1} \right)$$

$$= \frac{e^2}{2\pi \hbar} \sum_{n=0}^{n_{\text{max}}^2} \frac{1}{l_B^2} \left( ((n + \frac{1}{2}) + 6m_e^{-2} + l_{TB}^{-2})^{-1} - ((n + \frac{1}{2}) + 6m_e^{-2} + l_e^{-2})^{-1} \right)$$

$$+ \frac{3e^2}{2\pi \hbar} \sum_{n=0}^{n_{\text{max}}^2} \frac{1}{l_B^2} \left( ((n + \frac{1}{2}) + 4l_{as}^{-2} + 2l_m^{-2} + l_e^{-2})^{-1} - ((n + \frac{1}{2}) + 4l_{as}^{-2} + 2l_m^{-2} + l_{TB}^{-2})^{-1} \right)$$

where $l_B = (\frac{\hbar}{eB})^{1/2}$, $n_{\text{max}}$ is highest Landaue level quantum no

For simplicity we use some approximations

$$(q_n^2 + 6m_e^{-2} + l_{TB}^{-2})^{-1} \approx (q_n^2 + l_e^{-2})^{-1}$$

$$(q_n^2 + 4l_{as}^{-2} + 2l_m^{-2} + l_e^{-2})^{-1} \approx (q_n^2 + l_{as}^{-2} + l_e^{-2})^{-1}$$

To evaluate summation we use identity of Digamma function $\psi(x) = \frac{\Gamma'(x)}{\Gamma(x)}$ where prime denote derivative.

$$\psi(x) = \psi(x) + \sum_{n=0}^{N_{\text{max}}} \frac{1}{x+n}$$

$$\sum_{n=0}^{N_{\text{max}}} \frac{1}{\frac{1}{2} + \frac{2l_B^2}{l_B} + n_{\text{max}}} = \psi \left( \frac{1}{2} + \frac{2l_B^2}{l_B} + n_{\text{max}} + 1 \right) - \psi \left( \frac{1}{2} + \frac{2l_B^2}{l_B} \right)$$

For good conductor $n_{\text{max}}$ is very large number to find approximate value of $\psi \left( \frac{1}{2} + \frac{2l_B^2}{l_B} + n_{\text{max}} + 1 \right)$ we use an inequality

$$\ln(x) - \frac{1}{x} \leq \psi(x) \leq \ln(x) - \frac{1}{2x}$$

(for $x > 0$)

$$\lim_{x \to \infty} \ln(x) \frac{1}{x} \leq \psi(x) \leq \ln(x) - \frac{1}{2x} \leq \ln(\psi(x)) \leq \ln(x)$$

Above inequality only possible when

$$\lim_{x \to \infty} \psi(x) \longrightarrow \ln(x)$$

using above results in equation (10) it becomes

$$\Delta \sigma(B) = \frac{e^2}{\pi \hbar} \sum_{n=0}^{n_{\text{max}}^2} \left\{ \left( \frac{1}{2} + \frac{2l_B^2}{l_B} + \frac{l_B^2}{l_B} \right)^{-1} - \left( \frac{1}{2} + \frac{2l_B^2}{l_B} + l_e^{-2} \right)^{-1} \right\}$$

$$- \frac{3e^2}{\pi \hbar} \sum_{n=0}^{n_{\text{max}}^2} \left\{ \left( \frac{1}{2} + \frac{2l_B^2}{l_B} + \frac{l_B^2}{l_B} \right)^{-1} - \left( \frac{1}{2} + \frac{2l_B^2}{l_B} + \frac{l_B^2}{l_B} \right)^{-1} \right\}$$

$$= \frac{e^2}{2\pi \hbar} \left\{ - \frac{1}{2} \psi \left( \frac{1}{2} + \frac{2l_B^2}{l_B} + \frac{l_B^2}{l_B} \right) + \frac{3}{2} \ln \left( \frac{2l_B^2}{l_B} + \frac{l_B^2}{l_B} \right) - \psi \left( \frac{1}{2} + \frac{2l_B^2}{l_B} + l_e^{-2} \right) + \psi \left( \frac{1}{2} + \frac{2l_B^2}{l_B} + \frac{l_B^2}{l_B} \right) \right\}$$

$$+ \frac{3}{2} \psi \left( \frac{1}{2} + \frac{2l_B^2}{l_B} + \frac{l_B^2}{l_B} \right) - \ln \left( \frac{2l_B^2}{l_B} + \frac{l_B^2}{l_B} \right)$$

(13)
For some system if magnetic moment scattering is very weak $l_m \to \infty$ with this approximation equation (13) becomes

$$\Delta \sigma(B) = \frac{e^2}{\pi \hbar} \left[ -\frac{1}{2} \psi \left( \frac{1}{2} + \frac{B_s}{B} \right) - \frac{1}{2} \ln \left( \frac{B_s}{B} \right) - \frac{1}{2} \psi \left( \frac{1}{2} + \frac{B_0}{B} \right) + \frac{1}{2} \ln \left( \frac{B_0}{B} \right) \right]$$

$$+ \frac{3}{2} \psi \left( \frac{1}{2} + \frac{4r_B^2}{12a^2 + \frac{B_s}{B}} - \ln \left( \frac{4r_B^2}{12a^2 + \frac{B_s}{B}} \right) \right)$$

(14)

Now we able to reproduce explicitly mostly used for experimental reported equ for magneto conductance which is given by

$$\Delta \sigma(B) = \frac{e^2}{\pi \hbar} \left[ -\psi \left( \frac{1}{2} + \frac{B_s}{B} \right) + \ln \left( \frac{B_s}{B} \right) - \frac{1}{2} \psi \left( \frac{1}{2} + \frac{B_0}{B} \right) + \frac{1}{2} \ln \left( \frac{B_0}{B} \right) \right]$$

$$+ \frac{3}{2} \psi \left( \frac{B_{so} + B_s}{B} \right) - \frac{3}{2} \ln \left( \frac{B_{so} + B_s}{B} \right)$$

(15)

where $B_i = \frac{h}{2e}$ and $\phi$ phase de-coherent event by inelastic scattering which is identical with Thouless inelastic scattering in our discussion.

V. COMPUTATIONAL DETAILS

To explain the experimentally observed metal-insulator-transition (MIT) from the band theory perspective, we have carried out electronic structure calculations on bulk CCRO as well as on 4 u.c. and 2 u.c. CCRO films grown on (001) oriented LAO substrate. The structural design of the films are discussed in the next section. The results presented here are obtained from DFT calculations using the Quantum ESPRESSO simulation software [5]. Exchange-correlation potential is approximated through the Perdew-Burke-Ernzerhof general gradient approximation (PBE-GGA) functional [8]. The kinetic energy cut-off is set as 30 Ry. The Brillouin zone integration is carried out using the tetrahedral method through a Monkhorst-Pack k-point grid [9]. While a 4×4×1 k-mesh is considered for the structural relaxation, a denser k-mesh of 8×8×1 is used for the electronic structure calculations. The convergence criterion for self-consistent energy is taken to be $10^{-5}$ Ry. To account for the strong correlation present in Cu and Ru-d states, we have performed GGA+U calculations with $U = 2, 4, 6$ and $8$ eV. To examine whether the LAO substrate affects the electronic structure of the CCRO films through charger transfer and chemical bonding other than the epitaxial strain, the 2 u.c. and 4 u.c. films, without substrate but with equivalent epitaxial strain, are also examined.

VI. STRUCTURAL PROPERTIES

As suggested by the experiment, we have studied the electronic structures for 2 u.c. (∼1.5 nm) and 4 u.c. (∼3 nm) CCRO films to understand the dimensionality driven MIT. The CCRO thin films are grown along z-direction on 2×2×5 LAO substrate. 5 layers of LAO is found to be sufficient to form a substrate. The bottom three layers were fixed while the upper two layers were allowed to structurally adjust with the film. To consider the strain induced by the substrate, the in-plane lattice parameters of the modelled films are kept as twice of $a_{LAO}$ (as $a_{CCRO} \sim 2 \times a_{LAO}$, where $a =$ lattice parameter). The experimentally suggested out-of-plane lattice parameters are considered for electronic structure calculations of the films. A 15 Å vacuum was placed on the upper layer of CCRO slab such that a film is designed. At the beginning, the films were modelled with LAO symmetry and the ions were relaxed to minimize the forces to lower than 0.025 eV/Å. The relaxed 4 u.c. and 2 u.c. films and bulk CCRO are shown in Fig. S3(a), (b) and (c), respectively. As expected the structure becomes locally more distorted with reduced thickness. This is illustrated through the distortion of the RuO$_6$ octahedral complex (measured by bond length and bond angles), tilting of the neighboring octahedra (Fig. S3(d)) and shifting of the Cu atoms from the Ca-Cu-O plane (Fig. S4(e)).
Fig. S3: The relaxed structures of CCRO in (a) 4 u.c. and (b) 2 u.c. films grown on the LAO substrate, and in (c) bulk. To compare the local structural distortions as one moves from the bulk to film, the distortions of the RuO$_6$ octahedra, the tilting angle among the neighboring octahedra, and displacement of the Cu ions are shown.

Fig. S4: Band structure and densities of states (DOS) for bulk CCRO obtained with $U = 0$ and 6 eV.

VII. ELECTRONIC STRUCTURE

A. Strong Correlation effect in bulk CCRO

The bulk CCRO is a non-magnetic metal [9] as can be inferred from the band structure, shown in Fig. S4(a) and obtained within GGA, where a set of dispersive bands crosses the Fermi level. These partially occupied bands are
dominated by the delocalized Ru-d states as can be seen from the partial DOS plot shown in Fig.S4(c). The results are in agreement with the earlier reports [10]. The GGA+U ($U = 6$ eV) band structure (Fig.S4(b), (d)) are found to be no different which suggests that the compound is not a strongly correlated oxide.

B. Electronic structure of 4 and 2 u.c. thick CCRO films

![DOS plots showing the evolution of electronic structures](image)

**Fig.S 5:** The total, Cu-d and Ru-d DOS for the 4 u.c. CCRO film for different U values.

The evolution of electronic structures of 4 u.c. thick CCRO film with U is shown through the total and partial DOS in Fig.S5. With U, we find the gradual formation of Ru-d subbands on either side of the Fermi level ($E_F$). As a consequence, the net DOS at $E_F$ decreases with U. However, it is around 2 states per formula unit which is reasonably

![Orbital projected DOS plots](image)

**Fig.S 6:** The orbital projected DOS of Cu-d and Ru-d states in 2 u.c. CCRO films as a function of U.
Fig. S7: The total, Cu-d and Ru-d DOS for 4 u.c. and 2 u.c. thick films without substrate calculated with GGA+U method for U = 6 eV. The in-plane lattice parameters are kept as 2 × aLAO to consider the strain effects in the calculations. The experimental out-of-plane lattice parameters for 4 and 2 u.c. films are used.

less as compared to that of bulk (∼6 states at EF, Fig.S4(c-d)). This implies that the conductivity is significantly reduced as we move from the bulk to 4 u.c. film. In the case of 2 u.c. film, the number of states at EF falls below 1 (see Fig.4 of the main text) to induce an weak insulating behavior. To further examine the electronic structure of the 2 u.c. film as a function of U, in Fig.S6, we have shown the Cu-d and Ru-d orbital projected DOS for U = 0, 2, 4, and 6 eV. Within the independent electron framework (U = 0), we find that each of the five Ru-d orbitals occupy the EF, while Cu-d states create a small gap. As U increases, the onsite repulsion increases to create two subbands (lower and upper Hubbard bands; LHB, UHB) out of the Ru-d states. For U = 6 eV, the LHB and UHB are well separated to open a gap at the EF. However, some reminiscent states, arising out of the other elements of CCRO, lies at EF (see Fig.4 of the main text). These reminiscent states have minimal effect on the conductivity behavior of the film.

To ensure that the suggested MIT is not out of charge transfer and chemical bonding with the substrate, in Fig.S8, we have shown the total and partial DOS for the freestanding 4 and 2 u.c. films. They have a great resemblance to the ones grown on the substrate (see Fig.S7 and Fig.4 of the main text).

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