Electrically Driven Reversible Insulator-Metal Phase Transition in Ca_2RuO_4

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Introduction: Insulator-metal transitions (IMTs) are the subject of intense fundamental and applied research including their potential applications in electronic devices like coupled relaxation oscillators [1], neuromorphic devices [2], Phase FETs [3], and RF switches [4]. A key requirement for practical device application of IMT materials is that the IMT temperature (T_{IMT}) should be greater than 358 K (85C) which is the operating temperature of electronic chips (Fig. 1). In this work, we investigate the electrically induced IMT in epitaxially grown 0.3% tensile strained Ca_2RuO_4 thin films wherein strain engineering increases the transition temperature (T_{IMT}) to more than 550K from a bulk value of ~357K (ΔT_{IMT} >190K). Using systematic DC and transient I-V measurements, we show that the origin of the electrically induced IMT in Ca_2RuO_4 is current induced self-heating.

Experiment: Ca_2RuO_4 which belongs to the Ruddlesden-Popper series ($Ca_{n+1}Ru_nO_{3n+1}$ with n=1) exhibits an IMT at 357K (bulk) accompanied by an abrupt change in resistivity up to ~22x [5]. To further increase the T_{IMT} beyond 357K, we grow 20nm tensile strained Ca_2RuO_4 thin films on (110) $NdGaO_3$ using molecular beam epitaxy. The epitaxial growth of Ca_2RuO_4 on (110) $NdGaO_3$ induces a tensile strain of 0.3%, and increases T_{IMT} > 550K (maximum temperature range of the measurement setup) as shown in Fig. 2; the X-Ray Diffraction spectrum is shown in Fig. 3. This high value of T_{IMT} meets the temperature requirement for chip operation. The DC I-V characteristics of the two-terminal Ca_2RuO_4 devices shown in Fig. 4 exhibit non-linear behavior associated with the reduction in resistance across the IMT. While the voltage-mode I-V measurement shows an abrupt transformation in current associated with the IMT (along with hysteresis), the current-mode measurement exhibits a continuous negative differential resistance (NDR) across the phase transition, with no hysteresis. The abrupt current jump and hysteresis observed in the voltage-mode (in contrast to the current-mode) can be attributed to the additional joule heating (thermal runaway) that occurs when the resistance of the Ca_2RuO_4 device reduces across the IMT; no additional joule heating due to resistance reduction occurs in the current mode. The temperature dependent I-V characteristics shown in Fig. 5 reveal that the IMT can be electrically induced in Ca_2RuO_4 even at 373K (100C); and the evolution of the switching voltage and critical current associated with the IMT as a function of temperature is shown in Fig. 6.

To investigate the origin of the electrically induced IMT in Ca_2RuO_4, we perform transient I-V characterization using the setup shown in Fig. 7. Triangular ramp pulses with a peak amplitude of 8V and a pulse width (τ) ranging from 5ms to 1μs are applied, and the output voltage (V_{out}) is measured across the series resistance R_S (= 680 Ω). Figure 8 shows the evolution of the output voltage V_{out} for τ =1 ms, 100μs, and 1μs. It can be observed that non-linearity in the output, and consequently the peak output voltage V_{peak} decreases with τ (Fig. 10) indicating an incomplete IMT for shorter pulses. In fact, the absence of non-linearity in the output for τ=1 μs indicates the complete absence of the IMT. The corresponding I-V characteristics for the transient response shown in Fig. 9 also reflect the absence (or the incomplete nature) of the IMT process in Ca_2RuO_4 at shorter pulse widths (τ).

The strong sensitivity of the resistance non-linearity (induced by the IMT) to the time period τ of the applied pulse implies that the phase transition in Ca_2RuO_4 is electro-thermal in nature [6], and is driven by current induced self-heating of the Ca_2RuO_4 channel. Figure 11 shows the input energy supplied to the two-terminal device as function of τ. With reducing τ, the input energy (that gets converted to heat) also reduces, causing insufficient self-heating to initiate the IMT (e.g. τ=1 μs). Further, we note that the peak electric field across the device (corresponding to V_{in}=8 V) almost remains constant further suggesting that the IMT in Ca_2RuO_4 is not purely driven by the electric-field.

Conclusion: In summary, we have investigated the electrically induced IMT in Ca_2RuO_4 thin films whose transition temperature has been increased by >190 K (T_{IMT} > 550K) using epitaxial strain engineering. We show using DC and transient I-V measurements that the electrically induced phase transition is electro-thermal in nature, and is driven by current induced self-heating.

References: [1] N. Shukla *et al.* Sci. Rep., 4, 4964 (2014); [2] A. Sharma, VLSI 2015; [3] N. Shukla *et al.* Nat. Comm., 6 7812 (2015); [4] H. Madan *et al.* IEDM 2015; [5] T. Qi JSSC 184 (2011) [6] N. Shukla APL 105, 012108 (2014) [7] Yoon *et al.* Applied Materials & Interfaces, 8, 2280 (2016).

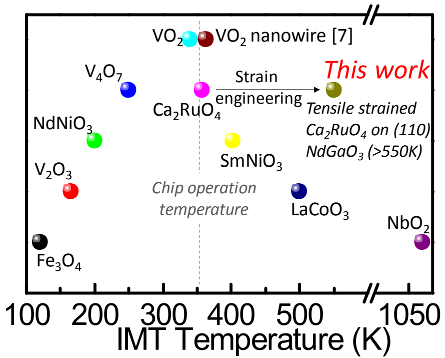

Fig. 1| Transition temperature (T_{IMT}) for various IMT materials. Practical device application requires T_{IMT} >358K (85C).

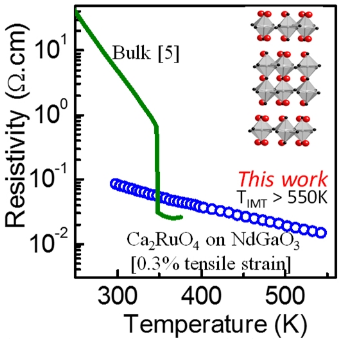

Fig. 2| Typical Resistivity vs. temperature characteristics for Ca$_2$RuO$_4$ on (110) NdGaO$_3$ (blue). Strain shifts T_{IMT}> 550K.

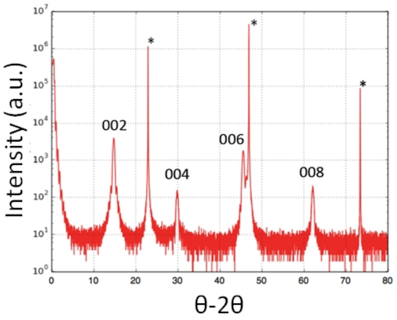

Fig. 3| XRD spectrum of the epitaxially grown Ca$_2$RuO$_4$ films grown on (110) NdGaO$_3$ (0.3% tensile strain).

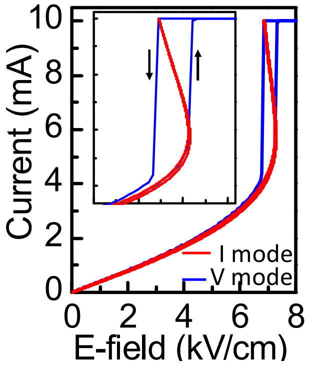

Fig. 4| I-mode and V-mode two-terminal I-V characteristics showing the IMT in Ca$_2$RuO$_4$. L=15 µm; W=40µm; T= 303K

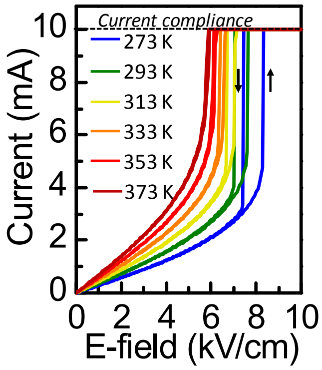

Fig. 5| Temperature dependent I-V characteristics showing the IMT in Ca$_2$RuO$_4$. L=15 µm; W=40µm.

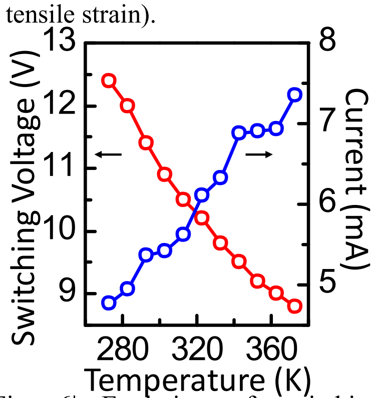

Fig. 6| Evolution of switching voltage and critical switching current for the IMT with temperature.

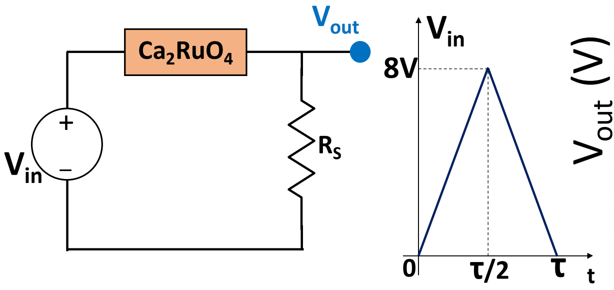

Fig. 7| Schematic of the pulse measurement setup and the input pulse. V_{out} is measured across R_S (=680 Ω). L=750 nm; W=2µm.

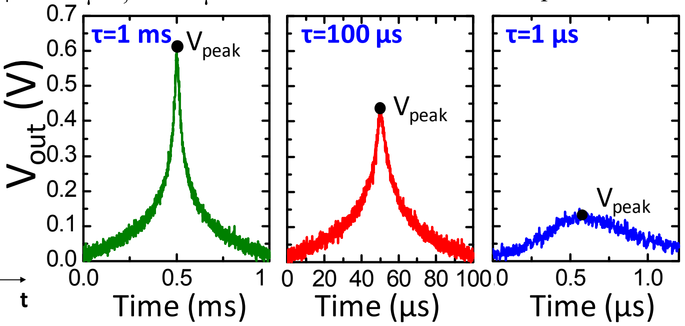

Fig. 8| Output voltage (V_{out}) as a function of pulse width (τ =1ms, 100 µs, 1 µs). The non-linear voltage evolution associated with IMT is not observed at smaller pulse widths.

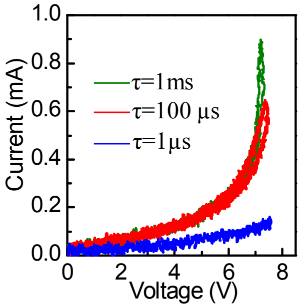

Fig. 9| Transient I-V characteristics as a function of the pulse width (τ).

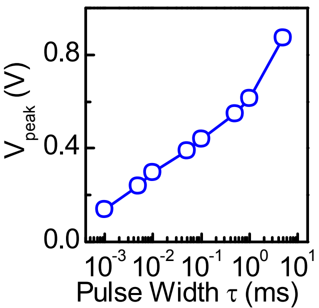

Fig. 10| Evolution of V_{peak} with pulse width (τ).

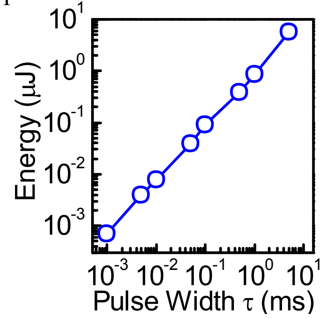

Fig. 11| Input energy supplied to the device as a function of τ. Reducing the input energy (thermal) fails to initiate the IMT.