Electrically Driven Reversible Insulator-Metal Phase Transition in Ca$_2$RuO$_4$

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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 (85°C) 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$_2$RuO$_4$ thin films wherein strain engineering increases the transition temperature ($T_{IMT}$) to more than 550K from a bulk value of ~357K ($\Delta T_{IMT} >190$K). Using systematic DC and transient I-V measurements, we show that the origin of the electrically induced IMT in Ca$_2$RuO$_4$ is current induced self-heating.

Experiment: Ca$_2$RuO$_4$ which belongs to the Ruddlesden-Popper series (Ca$_{n+1}$Ru$_n$O$_{3n+1}$ 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$_2$RuO$_4$ thin films on (110) NdGaO$_3$ using molecular beam epitaxy. The epitaxial growth of Ca$_2$RuO$_4$ 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$_2$RuO$_4$ 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$_2$RuO$_4$ 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$_2$RuO$_4$ even at 373K (100°C), 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$_2$RuO$_4$, 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 ($\tau$) ranging from 5ms to 1µs are applied, and the output voltage ($V_{out}$) is measured across the series resistance $R_S$ ( 680 $\Omega$). Figure 8 shows the evolution of the output voltage $V_{out}$ for $\tau$ = 1 ms, 100µs, and 1µs. It can be observed that non-linearity in the output, and consequently peak output voltage $V_{peak}$ decreases with $\tau$ (Fig. 10) indicating an incomplete IMT for shorter pulses. In fact, the absence of non-linearity in the output for $\tau$=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$_2$RuO$_4$ at shorter pulse widths ($\tau$).

The strong sensitivity of the resistance non-linearity (induced by the IMT) to the time period $\tau$ of the applied pulse implies that the phase transition in Ca$_2$RuO$_4$ is electro-thermal in nature [6], and is driven by current induced self-heating of the Ca$_2$RuO$_4$ channel. Figure 11 shows the input energy supplied to the two-terminal device as function of $\tau$. With reducing $\tau$, the input energy (that gets converted to heat) also reduces, causing insufficient self-heating to initiate the IMT (e.g. $\tau$=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$_2$RuO$_4$ is not purely driven by the electric-field.

Conclusion: In summary, we have investigated the electrically induced IMT in Ca$_2$RuO$_4$ 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.

This work

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

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

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. $V_{\text{out}}$ is measured across $R_{S} (= 680 \Omega)$. L=750 nm; W=2µm.

Fig. 8 | Output voltage ($V_{\text{out}}$) as a function of pulse width ($\tau = 1ms, 100 \mu s, 1 \mu s$). 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 ($\tau$).

Fig. 10 | Evolution of $V_{\text{peak}}$ with pulse width ($\tau$).

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