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Electrically induced insulator to metal transition in epitaxial SmNiO$_3$ thin films

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We report on the electrically induced insulator to metal transition (IMT) in SmNiO$_3$ thin films grown on (001) LaAlO$_3$ by pulsed laser deposition. The behavior of the resistivity as a function of temperature suggests that the primary transport mechanism in the SmNiO$_3$ insulating state is dominated by Efros-Shklovskii variable range hopping (ES-VRH). Additionally, the magnetic transition in the insulating state of SmNiO$_3$ modifies the characteristics of the ES-VRH transport. Systematic DC and pulsed current-voltage measurements indicate that current-induced Joule heating is the fundamental mechanism driving the electrically induced IMT in SmNiO$_3$. These transport properties are explained in context of the IMT in SmNiO$_3$ being related to the strong electron-lattice coupling. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4890329]

Perovskite rare-earth nickelates (RNiO$_3$) exhibit a prototypical thermally driven insulator to metal transition (IMT) (except for bulk LaNiO$_3$, which is a metal at all temperatures).$^1$ This along with competing interactions involving the lattice, orbital, spin, and charge degrees of freedom, which may be responsible for the IMT, make this material system the object of intense fundamental and applied research interest.$^2$–$^11$ The IMT in the RNiO$_3$ system usually consists of a transition from a low temperature insulating phase to a high temperature metallic phase with a bad-metal character.$^12$ The phase diagram of RNiO$_3$ compounds is controlled by the radius of the R$^{3+}$ atom, which in turn determines the tilting angle of (NiO$_6$)$_{3^-}$ octahedra.$^{13,14}$ The IMT temperature ($T_{\text{IMT}}$) decreases as the ionic radius of the R$^{3+}$ atom increases.$^{15}$ Nickelates also exhibit magnetic ordering.$^{16,17}$ In the case of PrNiO$_3$ and NdNiO$_3$, the Neel temperature ($T_N$) of antiferromagnetic ordering coincides with the IMT temperature ($T_{\text{IMT}}$). For Sm and smaller rare-earth metal ions, $T_N < T_{\text{IMT}}$.

SmNiO$_3$ exhibits an IMT above room temperature ($T_{\text{IMT}} \sim 400 \text{ K}$) transitioning from a low temperature insulating phase to a high temperature metallic phase. In this Letter, the electrically driven IMT in two terminal SmNiO$_3$ thin film devices using DC and pulse mode current-voltage (I-V) measurements is described. The ability to induce an IMT via an electric field is relevant to realizing electronic applications.$^{18,19}$ The mechanism of the electrically driven phase transition was investigated by applying the driving stimulus (I, V) using different time scales (200 ns–10 s) as a function of temperature.

SmNiO$_3$ thin films with a thickness of 16 nm were epitaxially grown on polished LaAlO$_3$ (001) substrates (MTI Corp.) by a pulsed laser deposition (PLD) system (Neocera) using a 248 nm KrF excimer laser (Coherent). A polycrystalline SmNiO$_3$ target of 99.99% purity (Shanghai Daheng Optics) was used. During growth, the substrate’s nominal temperature was at 1023 K in 33.3 Pa dynamic pressure of flowing O$_2$ with a laser pulse repetition rate of 5 Hz and a laser fluence of approximately 2 J/cm$^2$. After growth, the films were cooled at 20 K/min under the same O$_2$ pressure. The growth rate was calibrated using x-ray reflectometry (XRR). The structural quality of the substrate and the film was verified in-situ by reflection high-energy electron diffraction (RHEED) and ex-situ by x-ray diffraction (XRD) on a four circle diffractometer. Two terminal devices with dimensions $L = 2 \mu$m; $W = 4 \mu$m were fabricated using standard photolithographic techniques. The width of the device was defined by the contact pad. Ti/Au (40 nm/50 nm) was used for the contact pads resulting in ohmic contacts.

Figure 1(a) shows RHEED images taken from an as-grown thin film. The streaky nature and in-plane symmetry of the patterns revealed that the film surface was single-crystalline. Based on the RHEED results and the XRD Φ-scans of SmNiO$_3$ (101) and LaAlO$_3$ (111) pseudo-cubic peaks (Fig. 1(d)), we determined that the in-plane orientation of the pseudo-cubic unit cell vectors in SmNiO$_3$ coincide with the pseudo-cubic lattice vectors of the LaAlO$_3$ substrate. The same out-of-plane pseudo-cubic orientation, SmNiO$_3$ [001] || LaAlO$_3$ [001], was confirmed by XRD patterns (Fig. 1(b)), yielding an out-of-plane pseudo-cubic lattice constant $a = 3.81 \text{ Å}$ for SmNiO$_3$ resulting in a −0.6% mismatch with the substrate. The corresponding XRD rocking curve full width at half maximum (FWHM) value was 0.61° for the (002) SmNiO$_3$ peak (Fig. 1(c)).

The temperature dependent resistivity $\rho(T)$ of the SmNiO$_3$ film is shown in Fig. 2(a). The large change in resistivity, of over three orders of magnitude between 100 K and 400 K, further corroborates the good film quality. This epitaxially strained SmNiO$_3$ film undergoes an IMT at 393 K as indicated by the minimum in the $|d\rho/dT|$ (Fig. 2(a)).

To understand the nature of transport in the insulating phase of SmNiO$_3$, we plot $\ln(\rho)$ versus $T^2$, where $z = -1$ corresponds to Arhenius-type thermally activated conduction
weak AF signal and the value of $T_{N}$ from the minimum of the signal to be 230 K for ceramic samples. The proximity of the inflection point of $M$($T$) as a function of temperature is shown in Fig. 2(a). During field-cooling from 325 K in an external magnetic field of $H_{o}$ = 0.1 T. A blank LaAlO$_3$ substrate was measured in a SQUID magnetometer (Quantum Design) in the metallic state ($T < 273$ K) and in the insulating state of SmNiO$_3$. The current then became non-linear (more pronounced at lower temperature) with inflection points between the two regimes corresponding to $T_{N}$. Well below the IMT ($T < 273$ K, 333 K in Fig. 3(a)), the current was linear at low fields indicative of range hopping (ES-VRH) (Fig. 2(c)) to the $T_{N}$. The weak AF signal and the value of $T_{N}$ determined in this way was consistent with previous measurements on SmNiO$_3$ bulk ceramic samples. The proximity of the inflection point ($T_{N}$) in the ES-VRH case; Fig. 2(c)) to the $T_{N}$, within uncertainty, along with the strong correlation effects known to exist in this material system suggests that the transport mechanism is dominated by ES-VRH. The two distinct regimes in the ES-VRH process separated by the inflection point (closely corresponding to $T_{N}$) imply that even though the transport process across this transition may not change fundamentally, the lattice distortions related to the magnetic transition could modify the ES-VRH (temperature independent) transport parameters. In the ES-VRH transport regime, $\rho \propto \exp(TE_{S}/2)$, where $TE_{S}$ is the characteristic Efros-Shklovskii temperature. The corresponding fits to the data shown in Fig. 2(c) yielded the following $TE_{S}$ values: $TE_{S,1}$ = 1.62 × 10$^4$ K (0.0594 K$^{-1/2}$ < $T_{S}^{-1/2}$ < 0.12 K$^{-1/2}$) and $TE_{S,2}$ = 2.65 × 10$^4$ K (0.0522 K$^{-1/2}$ < $T_{S}^{-1/2}$ < 0.0594 K$^{-1/2}$).

Next, we studied the electrically (DC) driven phase transition in SmNiO$_3$ (Fig. 3(a)) at different temperatures. Well below the IMT ($T = 273$ K, 333 K in Fig. 3(a)), the current-electric field characteristics were linear at low fields indicative of the insulating state of SmNiO$_3$. The current then became non-linear (more pronounced at lower temperature $T = 273$ K) at higher fields, indicating the initiation of the IMT in SmNiO$_3$. In the metallic state ($T = 403$ K), the current was linear until at high electric field, additional Joule heating resulted in increased resistance and therefore reduced current. To study
The hysteresis in the electrically induced IMT as a function of temperature (Fig. 3(d)) was reduced as $T$ approached $T_{IMT}$. To explain this observation, we assume that the IMT in SmNiO$_3$ is electro-thermally triggered. The electro-thermal nature of the IMT in SmNiO$_3$ will be confirmed in the following sections. The sample was initially at a temperature $T$ ($=$ ambient temperature). As the voltage was increased, joule heating due to the current flow increased the local temperature of the sample. Hence, the sample temperature in the metallic low resistance state was much higher than $T$. As the field was swept back, SmNiO$_3$ was at an elevated temperature, and hence, the effective “turn off” voltage to return to the insulating state was lower. The hysteresis is therefore proportional to $\Delta = T_{ON} - T$, where $T_{ON}$ is the temperature of the SmNiO$_3$ film in the metallic state. For $T \rightarrow T_{IMT}$, $\Delta$ is smaller, and hence, the hysteresis observed was also less pronounced. While the above observations strongly point to a current induced joule heating driven IMT in SmNiO$_3$, these alone may not be sufficient to qualify Joule heating as the fundamental mechanism driving the IMT (other mechanisms like field induced resistance switching proposed by Sugimoto et al.$^{23}$ may have similar hysteresis characteristics).

To confirm whether the IMT was electro-thermally triggered, pulsed $I$-$V$ measurements were performed to analyze the IMT response to electrical stimuli applied on a wide time scale (200 ns–10 s). Figure 4(a) shows the circuit schematic for the pulsed $I$-$V$ measurements consisting of a two-terminal SmNiO$_3$ device with a 50 $\Omega$ series resistor ($R_s$; input impedance of oscilloscope). Triangular ramp pulses (Fig. 4(a)) with a peak amplitude of 20 V and time period ($\tau$) ranging from 200 ns to 10 s were applied.$^{24}$ The output voltage ($V_{Rs}$) was measured across $R_s$. Figure 4(b) shows $V_{Rs}$ for $\tau = 10$ s, 5 ms, 700 $\mu$s, and 1 $\mu$s (all the outputs are not shown for brevity) at $T = 298$ K. Similar measurements were also performed at $T = 323$ K and 348 K. Two features are clearly evident in the output characteristics: (a) the inflection corresponding to the change in resistance triggered by the IMT decreases with decreasing pulse period. This implies that the IMT was incomplete for shorter pulses; and (b) the peak output voltage $V_{peak}$ (across $R_s$) reduced with decreasing pulse width indicating that the SmNiO$_3$ film progressively undergoes a smaller change in conductance for shorter pulses (Fig. 4(c)). The absence of non-linearity in the $\tau = 1$ $\mu$s pulse implied the absence of an IMT (Fig. 4(b)). The critical pulse times $\tau_{cr}$ below which no non-linearity was observed are $\tau_{cr} = 100$ $\mu$s ($T = 298$ K); $\approx 50$ $\mu$s ($T = 323$ K); $= 1$ $\mu$s ($T = 348$ K).

The strong sensitivity of the non-linear change in resistance (induced by the IMT) to the time period of the applied pulse confirms that the phase transition in SmNiO$_3$ is driven by current induced self-heating.$^{25-28}$ Figure 4(d) shows the energy supplied through the input pulse and non-linear change in resistivity (resulting from the IMT) quantified as $dI/dV$ as a function of $\tau$. It is evident that the $dI/dV$ (indicating change in resistance) decreased with shorter pulse periods and then became constant implying no IMT. This is because the input energy (to be converted to Joule heat) was reduced, causing insufficient self-heating to initiate the IMT. Further, it is important to note that the peak electric field across the device (corresponding to $V_{in} = 20$ V) remained nearly constant. Therefore, the possibility of the
IMT in SmNiO₃ being driven by a purely electric field effect is unlikely.

From the I-V characteristics (across the SmNiO₃ device), for the \( \tau = 10 \) s and \( \tau = 1 \) ms case (Fig. 4(e)), it is evident that the I-V characteristics for the \( 1 \) ms pulse were linear, indicating that the resistance was constant and no IMT was triggered despite of the peak input ramp voltage \((20 \) V) remaining the same. Unlike the \( \tau = 1 \) ms case, the \( \tau = 10 \) s case had a non-linear evolution of the resistance, indicative of the occurrence of an IMT.

We now discuss our results in the context of the nature of the IMT in SmNiO₃. The rare earth nickelates RNiO₃ are classified as charge transfer insulators according to the Zaanen-Sawatzky-Allen criteria for correlated oxides, with the insulating band-gap formed between the oxygen 2p and the Ni 3d bands. In case of the nickelates, the overlap of the lower Hubbard band with the oxygen p band depends on the angles of the Ni-O bonds. This so-called negative or zero charge transfer causes a strong dependence of the band gap on the Ni\(^{3+}\)-O\(^{2-}\) hybridization and its geometrical arrangement.

Recent studies have pointed out the importance of a two Ni-site distribution, between the more ionic NiI and the more covalent Ni2 sites with longer and shorter Ni-O bonds, respectively, which are present in the low temperature insulating phase, described within the monoclinic P2₁/n space group. Johnston et al. recently proposed that the IMT in nickelates originates from a strong coupling between the O ligand holes as charge carriers and the rocksalt-like lattice distortions related to the Ni1 and the Ni2 sites in distinct (NiO₆)\(^{3-}\) octahedra.

When SmNiO₃ is electrically driven, Joule self-heating results in a reduction of the lattice distortion in the insulating phase and in the corresponding band gap closure. The transport properties in the metallic state remain affected by the electron-phonon interactions. This was corroborated by Jaramillo et al. who observed that a mid-infrared peak corresponding to the bandgap-like feature in the insulating state persists as a Holstein polaronic side band in the metallic state. Mediated via electron-lattice interaction, gradual band structure transformation with increasing temperature results in a non-linear increase in the conductivity as observed experimentally. Further, with pulse I-V measurements, the increase in local temperature induced by Joule heating becomes progressively smaller, and therefore the non-linear increase in resistivity induced by the IMT diminishes.
In summary, we have investigated the IMT in epitaxial SmNiO$_3$ thin films grown on (001) LaAlO$_3$. Temperature dependent resistivity measurements indicate that Efros-Shklovskii VRH is likely to be the dominant transport mechanism in the insulating state of SmNiO$_3$. Further, we explore the electrically driven IMT in SmNiO$_3$ through DC and pulse I-V measurements over a large time scale elucidating the fundamental mechanism driving the IMT in SmNiO$_3$ to be current induced Joule self-heating.

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24The following time periods were used. $\tau = 200$ ns, 500 ns, 700 ns, 1$\mu$s, 5$\mu$s, 7$\mu$s, 10$\mu$s, 50$\mu$s, 70$\mu$s, 100$\mu$s, 500$\mu$s, 700$\mu$s, 1 ms, 5 ms, 7 ms, 10 ms, 50 ms, 100 ms, 500 ms, 700 ms, 130 ns, 10 s.