	Experiment title: Unravelling a photoinduced compressive lattice response in the canonical Mott insulator (V1-xCrx)2O3	**Experiment number**: MA-5554
Beamline: ID09	**Date of experiment**: from: September 6 to: September 12, 2023	**Date of report**: March 4, 2023
Shifts: 18	**Local contact(s)**: Matteo Levantino	*Received at ESRF:*

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Report:

Objectives. This proposal aimed at determining the nature of lattice strain, compressive *vs* tensile, following the ultrafast photoexcitation of hot carriers in the canonical Mott insulator $(V_{1-x}Cr_x)_2O_3$. Our recent µ-XRD mapping on $(V_{0.95}Cr_{0.05})_2O_3$ thin films after an electric field-driven insulator to metal transition indeed revealed a compressive lattice response, as shown in Figure 1(a-b). As electric field and light pulses are both prone to generate hot carriers, we expected that photoexcitation would also induce a compressive lattice response in $(V_{1-x}Cr_x)_2O_3$. Our preliminary pump-probe reflectivity study in $(V_{0.95}Cr_{0.05})_2O_3$ films is consistent with this view. Overall this study might challenge the paradigm by which photoexcitation of matter always leads to a thermally-driven expansion, and suggest that Mott physics can lead to a reverse compressive behavior. This study is part of the MOTT-IA project, which aims at creating the first hardware neural network based on the properties of Mott insulators.

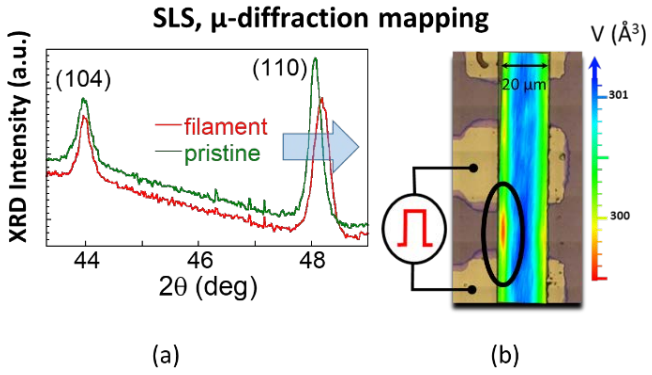

Figure.1 : (a-b) µ-XRD mapping experiment performed on a device made of $(V_{0.95}Cr_{0.05})_2O_3$ thin-film submitted to a Electric Mott Transition. (a) Comparison of µ-XRD diffractograms inside and outside the metallic filamentary path induced by electric pulse. (b) Map evidencing the unit cell volume contraction (shift of Bragg peaks towards high Q or equivalently to high diffraction angle 2θ) inside of the filament.

Experiments performed and main results

We have performed time resolved X-ray diffraction on four different $(V_{1-x}Cr_x)_2O_3$ thin films, whose positions in the phase diagram are indicated in Figure 2 (a):

(1) on a x= 0.05 sample (DET48, gap 0.15 eV, thickness 100 nm), using a pump energy at 2300 nm (0.5 eV). We measured temporal scans in the 0-1 ns range and fluence scans up to 29 mJ/cm², as shown in Figure 2(b-c). Contrary to what we expected, a photoinduced shift towards lower Q is observed for all the Bragg peaks, indicating a volume expansion.

(2) on a x = 0.12 sample (DET 30, gap ≈ 0.25 eV, thickness 100 nm), with two pump energies (0.5 eV and 1.55 eV). Our hope was that the gap of this sample (≈ 0.25 eV), closer to the pump energy of 0.5 eV compared to the x = 0.05 sample (gap ≈ 0.15 eV), would lead to better conditions to observe the compressive effect. However, Figure 2(d-e) shows that the shift of the Bragg peak is still towards lower *Q*, *i.e.* towards a volume expansion. Still, the smaller volume expansion observed at a pump energy of 0.5 eV compared to 1.55 eV at the same total fluence suggests the possibility of a competition with a compressive mechanism. Indeed both volume expansions should be similar if only a tensile mechanism was present.

(3) on a pure V_2O_3 sample (DET 44, thickness 116 nm), using the 1.55 eV pump laser. As expected in a metallic phase, the photoinduced volume change is expansive (tensile), with a much larger effect compared to the Mott insulators (PI) samples DET 48 (x = 0.05) and DET 30 (x = 0.12).

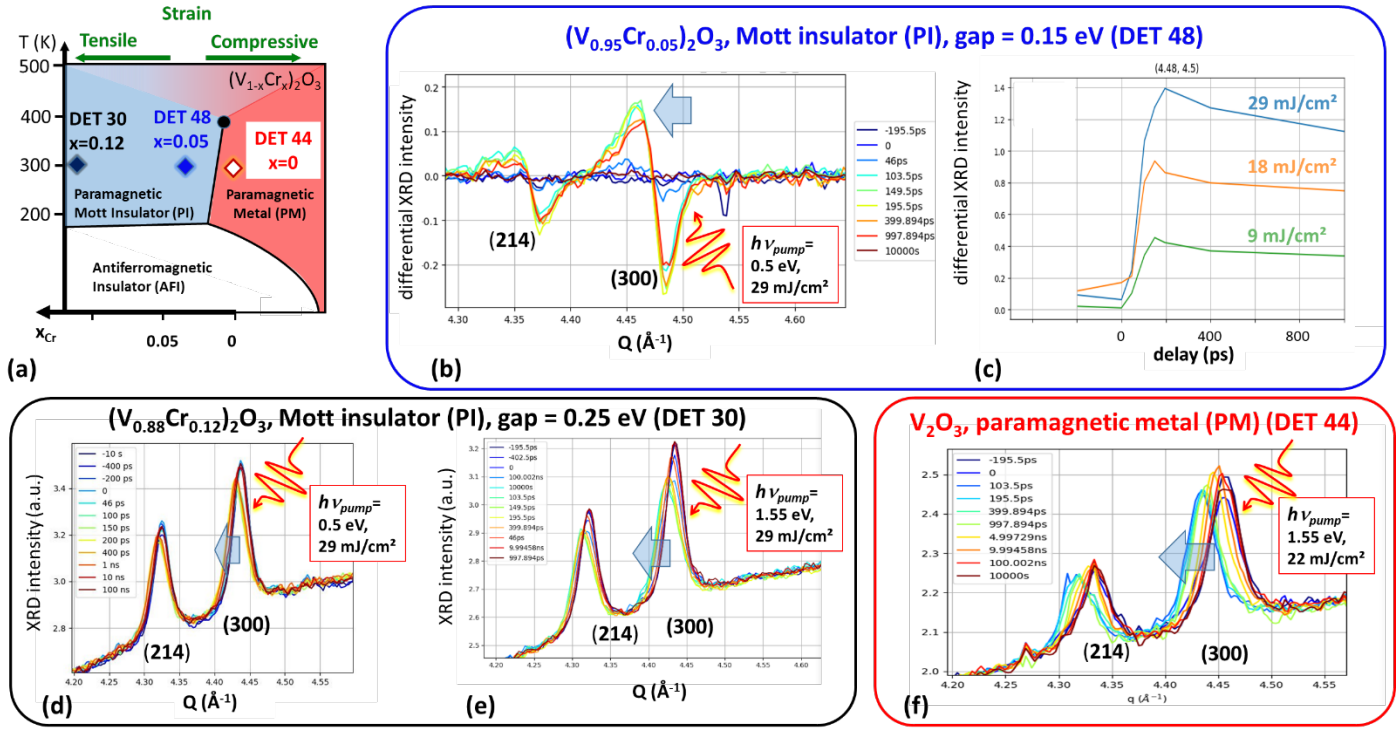

Figure.2 : (a) temperature vs strain (top horizontal axis) and composition x (bottom horizontal axis of the $(V_{1-x}Cr_x)_2O_3$ system. The position in the phase diagram of the three thin films measured during this run are indicated as diamond symbols. (b-c) x = 0.05 sample (DET 48, PI phase). Differential XRD pattern measured with the 0.5 eV pump laser at 29 mJ/cm² at various delays (b) and associated time dependence (c). (d-e) x = 0.12 sample (DET 30, PI phase). XRD pattern measured with a 0.5 eV pump with a 29 mJ/cm² fluence at various time (d). Same with a 1.55 eV pump (e). (f) x = 0 sample (DET 44, PM phase). As expected for a metal, a rather strong volume expansion is observed due to a thermoelastic effect.

Overall, the results obtained on both samples in the PI phase are surprising. We have indeed recently detected a strong ($\Delta V/V$ = -1.5 %) and non-thermal photoinduced compressive effect in a V_2O_3 film in the AFI phase at low T (10 K). A possible explanation is that the total photoinduced stress results from the competition between two effects, one compressive and one tensile. As the tensile effect is proportional to the thermal expansion coefficient, it is expected to be larger at 300 K (where the above results were obtained) than at 10 K (where the compressive effect was detected). Moreover the laser energy used in this ID09 experiments (0.5 eV) is two or three time larger than the gap of the measured samples (≈0.15 eV for x = 0.05 and ≈0.25 eV for x = 0.12). The pump laser energy in excess with respect to the gap in transformed into heat, and hence to a tensile effect.

Our next session at ID 09 in May 2023 should help to clarify the situation, since we will measure samples with a gap much closer to the pump laser energy available at ID09 (0.5 eV), either by working at low temperature or using V2O3 sample with much higher chromium content.