ESRF	**Experiment title:** Mott transition in NiO under pressure	**Experiment number**: HC-811
Beamline: ID09	**Date of experiment**: from: 24 June 2013 at 08:00 to: 27 June 2013 at 08:00	**Date of report**: 03.09.2014
Shifts: 6	**Local contact(s)**: Michael Hanfland	*Received at ESRF:*

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Report:

Many of the insulating transition metal and rare earth compounds exhibit localized magnetic moments that result from strong electron correlations and the partial filling of the 3d and 4f states, respectively. The physics of the magnetic ordering of these substances and the corresponding ordering temperatures are defined by various exchange and super-exchange mechanisms. The application of hydrostatic pressure results in increased spatial overlap of the wave-function, which causes band shifts, overlap, broadening, and stronger band hybridization. These effects in turn can lead to delocalization of the electronic wave functions and, thus, to the concurrent metallization and collapse of magnetic moments in the narrow pressure range [1]. On the other hand, such transition can be explained by the change of the crystal-field splitting resulting in the high-spin to low-spin transition of the transition metal [2].

The transition-metal oxides, as FeO, MnO, and NiO are the typical Mott insulators [1], which are actively used as model systems for the theoretical calculations [3]. The metallization and magnetic collapse were observed [4,5,6,7,8,9] experimentally in FeO and MnO at the pressures exceeding 70 GPa and 100 GPa, respectively. Different methods have been used to characterize the materials around this pressure range and to describe their electronic, structural, and magnetic properties. However, the origin of the observed transition is still under debates [2,9]. As compare to FeO and MnO, the investigations of the NiO, which was treated by Mott as prototype "Mott" insulator, are scanty. The theoretically predicted phase transition pressure is above 200 GPa [3]. The number of experimental methods that can be applied at this pressure range was very limited up to now. However, recent improvements of the high-pressure techniques allows for investigation of 200-300 GPa range [10,11]. Recently, the long-sought insulator-metal transition in nickel oxide has been experimentally found at ~240 GPa [10], where drop of resistivity by 3 orders of magnitude has been observed (see Fig.1).

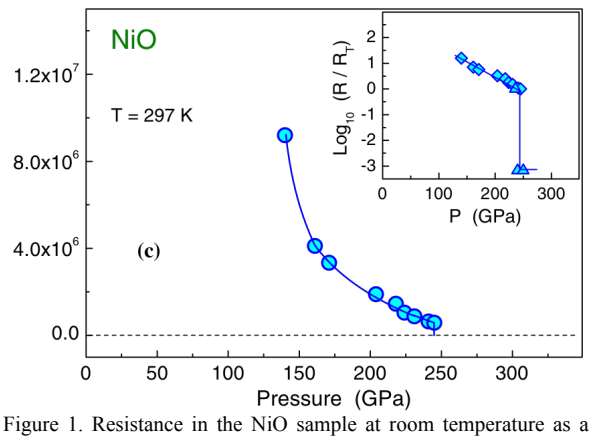

Figure 1. Resistance in the NiO sample at room temperature as a function of pressure. Inset shows drop of resistance at insulator-metal transition [8].

There exist several possible cases of relationship between electronic and structural transitions [12]: (1) "pure" electronic transition without any appreciable structural alterations (Mott-Hubbard transition in NiI_2); (2) A more general case when the electronic/magnetic transitions corroborate with a substantial structural transformation, usually a first-order

isostructural phase transition (Mott transition in FeI_2 and $FeCl_2$, high spin – low spin in $RFeO_3$, quenching of orbital term in FeI_2, $FeCl_2$); (3) Structural phase transitions which induce an electronic transition (*e.g.* Fe_2O_3). The single crystal x-ray diffraction is an ideal method for studying any kind of structural transitions.

Within the beam time allocated for proposal HC-811 we performed a combined single crystal and powder X-ray diffraction study of NiO structure behaviour under pressure.

We performed a single crystal measurments from 0 to 59 GPa and powder measurments from 120 to 220 GPa. From Single crystal study we manage to extract a high quality EOS for NiO to which our powder data are fitting nicely. The data extracted from powder run indicate absence on any structural transformation up to 220 GPa. We had also perfomed invetigation of samples left from spectroscopy experiment HC-830. After HC-830 we had 3 pressuriesed DACs left, compressed to 150, 240, 280 GPa representevly. Unfortunatly, we discovered the in all of the DAC pressure drope below 200 GPa due to the diamonds failure over time that separated to expereriments. Therefore due to time separation of the two beamtimes, we didn't manage to obtaine the x-ray data above the transitin pressure with samples investigate by Nuclear Forward Scattering erlier.

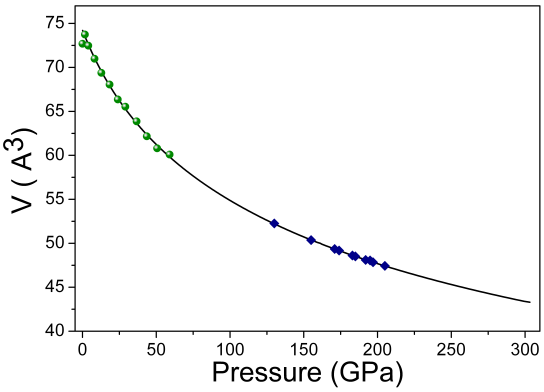

Figure 2. Pressure dependence of the volume of NiO extracted fro single crystal XRD measurements (olive circles) and from powder XR measurements (Blue diamonds). The black curve is EOS extracted fro single crystal measurements,

In summary, during the beamtime we performed complex X-ray diffraction investigation of NiO strucural behaviour under pressure up to 220 GPa. In this pressure range we didn't observe any sign of structural transofrmation that could be associeted with Mott Transition.

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