ESRF	**Experiment title:** Stress and structure of epitaxial CoO(111) layers on Ir(001)	**Experiment number**: SI-1757
Beamline: ID-03	**Date of experiment**: from: 01-Oct-2008 to: 08-Oct-2008	**Date of report**: 27. July 09
Shifts: 21	**Local contact(s)**: Dr. Roberto FELICI	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

SANDER, Dirk*

MEYERHEIM, Holger,*

TIAN, Zhen*

KIRSCHNER, Jürgen

All at: Max Planck Institute of Microstructure Physics, Halle, Germany

FELICI, Roberto, ESRF, Beamline ID-03

Report:

The growth of monolayer thin CoO(111) films on Ir(001) has created significant interest recently. This interest is partly due to the fact that the growth of CoO(111) in its bulk rocksalt strcuture would lead to adjacent Co^{++} and O^{--} layers with a correspondingly large repulsive Coulomb-interaction. This aspect is of large fundamental interest because either a Coulomb-driven structural change might be anticipated, or the limited thickness of two monolayers might lead to an acceptable low level of repulsive interaction, rendering the rocksalt strcuture possible in the monolayer regime. However, the results of our studies at beamline ID-03 clearly demonstrate that the rocksalt structure is not observed, even in two monolayer thin CoO(111) films.

Previous structural invenstigations by low energy electron diffraction (LEED) [1] have identified a c-10x2 diffraction pattern in monolayer thin CoO(111) films, but no in-depth structaural analysis has been presented. Our own LEED and stress measurements on 2 ML CoO(111) on Ir(001) have also identified the c-(10x2) diffraction pattern [2]. Remarkably, a *tensile* stress of the CoO(111) monolayers, in quantitative agreement with a misfit induced stress, has been detected. We measure a tensile surface stress change of $\tau_{exp} = +2.1$ N/m upon formation of 2 ML CoO(111)-c-10x2 on Ir(001). This value agrees quantitatively with the calculated domain-averaged misfit-induced stress of +2.05 N/m. This result of a tensile stress already disqualifies any structural model which is based on charged Co^{++} and O^{--} layers, where a large compressive stress, due to a repulsive Coulomb interactions within the layers, would have resulted. Thus, a structural relaxation could be anticipated, which would lead to a reduced Coulomb interaction within the layers. Our surface x-ray diffraction (SXRD) experiments during this beamtime provide strong support for the presence of a structural phase, which is characterized by a (local) rocksalt (RS)- followed by a Wurtzite (WZ)-type stacking sequence. We observe strong relaxations involving a reduction of the vertical spacing between

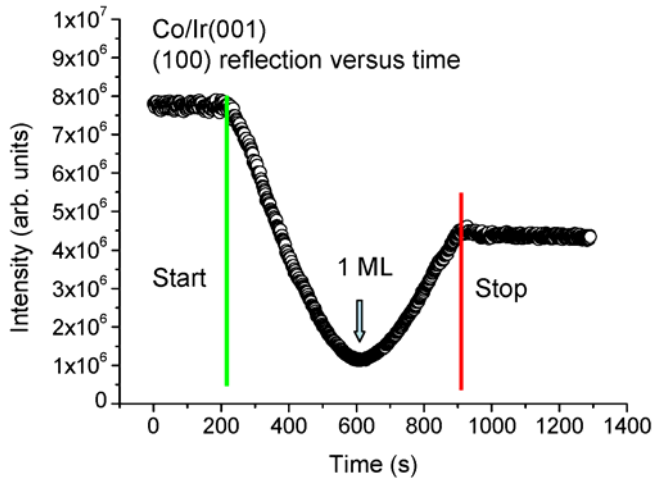

Fig. 1. Intensity of the (100) diffraction peak during deposition of 2 ML Co. The observation of a maximum at 910 s identifies the completion of the 2nd Co layer.

the atomic planes of cobalt- and oxygen-atoms from its CoO bulk value of 1.23Å to close to zero. Tentatively, this might be described as the formation of graphitic CoO layers. Both, relaxations and modifications of the layer stacking sequence are assumed to lead to a cancellation of the layerwise repulsive interraction, where also the metallization of the surface might play a role [1].

A prerequisite for a trustworthy structural analysis was the preparation of a CoO film of known thickness. To this end we have calibrated the Co source by SXRD, and Figure 1 shows a plot of the (100) antiphase intensity measured during Co deposition onto Ir(001) at 300 K. The first minimum at 600 s marks the completion of the first layer Co, the maximum at 910 s marks the completion of the second layer. The integrated Co ion flux during deposition served as a crosscheck for the Co deposition during CoO formation.

We prepared CoO(111) films containing the equivalent of 2 ML Co by deposition of Co at an elevated temperature of 600 K in an oxygen atmosphere of $2x10^{-6}$ mbar O_2. Figure 2 presents the sketch of the reciprocal lattice and some measured L-scans, together with a fit (blue curve) through the data points. The L-scans reveal intensities at the characteristic H K positions as inferred from the sketch of the c-10x2 structure on the reciprocal map.

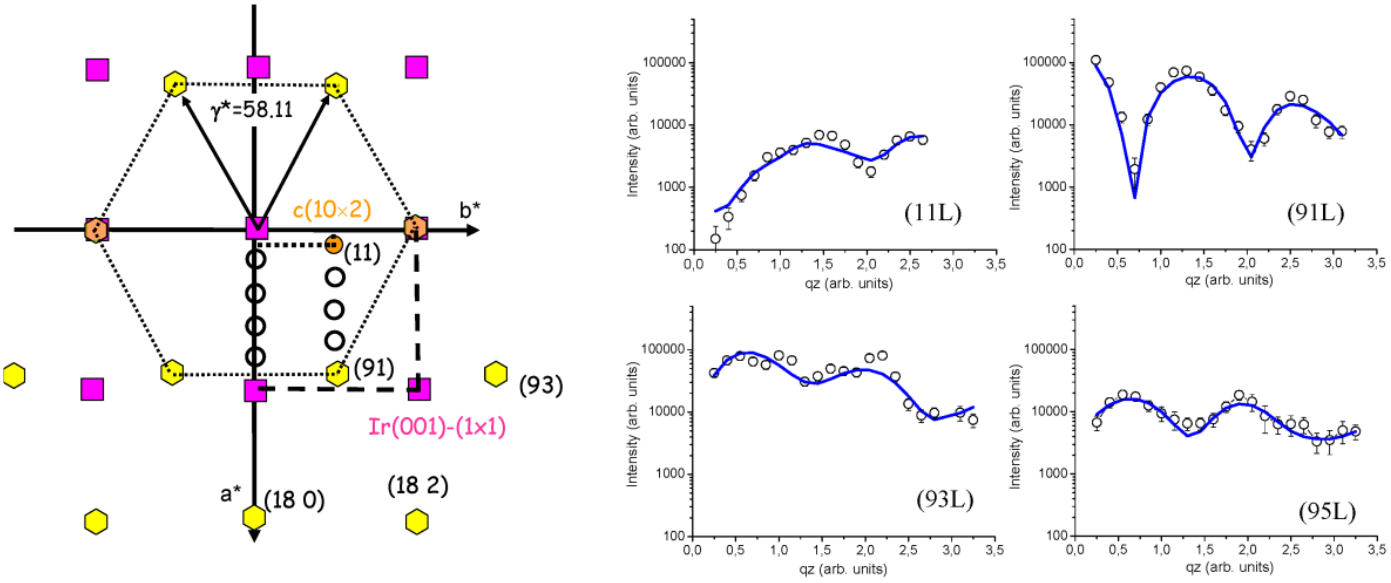

Fig. 2. Sketch of the reciprocal lattice of Ir(001) (squares) and of the locations of some c-10x2 related superstructure spots (circles and hexagons). Some of the H, K positions are lableled, where the L-scans, shown at right, were measured. The blue curve is a fit based on the structural model of Fig. 3.

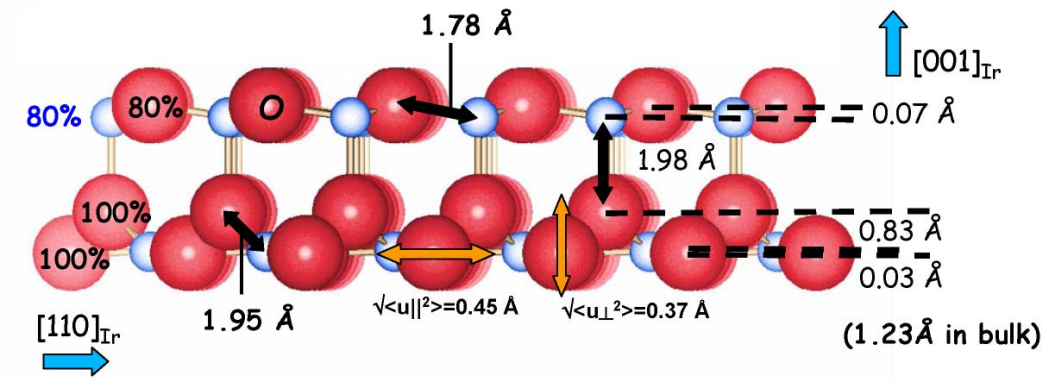

Fig. 3. Structure model of 2 ML CoO(111) on Ir(001). Oxygen and Cobalt atoms are shown as red (large) and blue (small) spheres, respectively. Main features are the presence of both, a relaxed RS and WZ type structure and strong vertical relaxations of Co and O towards each other in each layer. The oxygen atoms at the Ir interface at the bottom occupy along Ir[110] on-top, bridge, and on-top positions (not shown).

This result supports the finding of a c-10x2 structure of CoO on Ir(001). The L scans show pronounced modulations as a function of qz. This is ascribed to intereference of diffraction from different layers within the structure. Excellent fits could be obtained, in which the calculated intensities (blue curve) follow the experimental ones (points) over a dynamic range of more than three orders of magnitude.

Figure 3 reveals the structure model which is the basis of the fit to the data points of the L scans of Fig. 2. We find three oxygen layers and two cobalt layers. The atomic positions are arranged in a strongly relaxed RS stacking, followed by a WZ-type stacking along [001. Cobalt atoms are located on top of oxygen-atoms at a distance of 1.98 Å. The layers exhibit strong vertical (⊥) and lateral (||) modulations, which are described by by root mean squared (rms) displacement amplitudes of $\sqrt{<U_{||}>^2}=0.45$ and $\sqrt{<U_{\perp}>^2}=0.37$ Å, respectively. The modulations result form the accommodation of the oxide on the Ir(001) surface with a 9/10 coincidence along [110]. The detailed analysis reveals oxygen sites in a top-bridge-top sequence on Ir, and this variation of the oxygen position induces a rumpling within the layers. The Co atoms are positioned almost within the same layer, displaced vertically by only 0.03 Å. The same, almost in-plane, proximity of Co and O is also found at the top layer, where the displacement between the layers of Co and O amounts to 0.07 Å. Note, that a layer of O is introduced 0.83 Å above the 1st Co layer next to the Ir interface.

In conclusion, the preparation of 2 ML CoO in a c-10x2 structure on Ir(001) was successfully accomplished by Co deposition in an oxygen atmosphere at elevated temperature. The structure analysis of CoO on Ir(001) reveals a c-10x2 unit cell, where layers of Co and oxygen are only shifted very little (0.08 A) against eachother. RS-type and WZ-type structures coexist, and this result disqualifies the assumption of a simple RS type structure in 2 ML CoO on Ir(001). We speculate that the obserevd structural details might induce an almost complete quenching of the repulsive Coulomb interaction, which would have resulted from bulk-like RS structure. This finding offers an explanation of our stress measurements, where we have found no indications for repulsive intralayer interactions.

References:

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W. Meyer, D. Hock, K. Biedermann, M. Gubo, S. Müller, L. Hammer, and K. Heinz
Phys. Rev. Lett. **101**, 016103 (2008)
[2] Zhen Tian, PhD thesis, Martin-Luther-University Halle-Wittenberg, Germany, 2008.