Investigation of the orbital ordering in a La$_{2/3}$Sr$_{1/3}$MnO$_3$/BaTiO$_3$ multilayer

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We report here on the first results of x-ray diffraction experiments, aiming at investigating the orbital order induced by epitaxial strain in a multilayer composed of two transition metal oxides. As a matter of fact, a theoretical study proposes that in a heterostructure made of the repetition of 3 unit cells (UC) of La$_{2/3}$Sr$_{1/3}$MnO$_3$ (LSMO) and 3 UC of BaTiO$_3$ (BTO), epitaxial strain leads in the LSMO layers to an expansion of the upper and lower UC along the c-direction and therefore to a higher occupancy of the d–(3z$^2$ - r$^2$) orbital of the Mn$^{3+}$ ions, and to a compression of the inner UC leading to a higher occupancy of the d–(x$^2$ - y$^2$) orbital. Because of this orbital ordering, this system is supposed to exhibit a significantly increased Curie temperature of ferromagnetic ordering. [1]

In order to experimentally verify the theoretical predictions, we tried two types of x-ray diffraction experiments on beamline P09. We first performed a depth resolved investigation of the lattice distortions by high-energy x-ray diffraction, giving indirect information on the orbital ordering. Secondly, the orbital occupancy of the Mn$^{3+}$ ions was investigated by resonant scattering at the Mn K-edge.

The sample was prepared by molecular beam epitaxy and was characterized magnetically in Caen. The sample was then characterized structurally in Jülich by x-ray reflectometry and wide angle diffraction. Those measurements show the high structural quality of the sample. The bilayer thicknesses and the average lattice parameters of the different layers were determined. [2]

First, we performed non-resonant x-ray diffraction with the scattering wave vector (Q) parallel to the c crystallographic direction, using high energy (E = 20 keV) in order to reach large Q values. The scan, measured up to the (0 0 10) fundamental Bragg reflection is shown in Fig. 1. An analysis of those data should, in principle, allow us to determine the lattice parameters in each unit cell of each layer.

Fig. 1: (left) (00l) scan performed on P09 at E = 20 keV on a [LSMO/BTO]$_{27}$ multilayers. (right): Detail around the (001) fundamental Bragg peak of the multilayer. Red: data. Green: simulation
Second, we performed resonant x-ray scattering at the Mn K-edge. When the eg Mn orbitals show ordering, the atomic scattering factor of Mn becomes anisotropic near the Mn-K absorption edge. At P09 the synchrotron radiated x-beam is linearly polarized in the horizontal plane and the scattering plane is vertical. Since the expected ordering of the Mn eg orbitals does not lead to Bragg peaks at positions in reciprocal space different than the ones of the Bragg peaks induced by conventional charge scattering and that, therefore, the conventional charge scattering always contaminates the $\sigma\rightarrow\pi'$ channel (due to the fact that the scattering angle in the polarization analyzer is never exactly equal to 90°), a relatively new technique was used, namely the interference technique [3,4]. This technique counterbalances the contamination by subtraction of signals taken at two values of the angle $\varphi_A$ of the “analyzer and detector assembly” (that rotates in the plane normal to the scattered wavevector $\mathbf{k}'$) chosen symmetrically around the $\sigma\rightarrow\pi'$ channel for which channel $\varphi_A = -90°$. We measured those signals as a function of energy at $Q$ fixed to a certain superlattice Bragg peak and for several values of the azimuthal angle $\psi$ (a rotation angle around the scattering wave vector). Fig. 2 shows them measured at $\psi=30°$. The two measurements at $\varphi_A = -80°$ and $\varphi_A = -100°$ show qualitative differences around the Mn K-edge ($E=6555$ eV), a clear signature of the anisotropy of the atomic scattering factor of Mn! Unfortunately, those two signals do not coincide at energies away from the edge, preventing us to exploit their difference. This might be due to the fact that those signals were not integrated over the (very small) rocking curve of the Ge (100) analyzer crystal. However, this first test experiment is a clear indication that the Mn orbital ordering can be investigated by this technique!

![Graph](image)

Fig. 2: Energy scans at the $l=-1$ satellite peak of the (102) fundamental Bragg peak. The three scans were performed at the same azimuthal angle $\psi = 30°$ (sample rotation angle around the scattering vector counted relative to the (100) crystallographic direction) and at three different values of the analyzer angle $\varphi_A$.

References