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## Improved Structural Properties in Homogeneously Doped Sm<sub>0.4</sub>Ce<sub>0.6</sub>O<sub>2-6</sub> Epitaxial Thin Films: High Doping Effect on the Electronic Bands

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The study of ionic materials on nanometer scale is of great relevance for efficient miniaturized devices for energy applications. The epitaxial growth of thin films can be a valid route to tune the properties of the materials and thus obtain new degrees of freedom in materials design. High crystal quality  $Sm_xCe_{1-x}O_{2-\delta}$  (SDC) films were grown at a high doping level up to x = 0.4, thanks to the good lattice matching with the (110) oriented NdGaO<sub>3</sub> (NGO) substrate. Transmission electron microscopy measurements reported in Figure 1 demonstrate the ordered structural quality and absence of Sm segregation at the macroscopic and atomic level. Therefore, in epitaxial thin films, the homogeneous doping can be obtained even with the high dopant content not always approachable in bulk form, getting even an improvement of the structural properties. In situ spectroscopic measurements by X-ray photoemission and X-ray absorption show the O 2p band shift toward the Fermi level (see Figure 2), which can favor the oxygen exchange and vacancy formation on the surface when the Sm doping is increased to x = 0.4, in agreement with the results reported in Nan Yang et al. ACS Appl. Mater. Interfaces 2016, 8, 14613.



Fig. 1: HRTEM analysis of the Sm doped film with x = 0.4: (a) bright-field micrograph showing substrate and film. (b) FFT from the film (yellow) and substrate region (cyan). (c) SAED pattern of the film. (d) EDX analysis. (e) STEM ADF image, the rectangle marks the region chosen for EDX elemental mapping. (f-h) EDX elemental maps for Sm, Ce, and Nd. Scale bars in (e-h) are 10 nm.



Fig. 2: Valence band spectra in situ measured with 495 eV photon excitation energy for undoped CeO<sub>2</sub> and SDC with x = 0-0.4 in the energy range 0–5 eV. The colored regions point out the different contributions to the valence band obtained by fitting after subtracted a Shirley background. In the inset, sketch of the largest SDC40 crystallographic cell, almost unstrained when grown on the NGO substrate.



