Analysis of atomic-scale and electronic structures of epitaxial Mg$_x$Ni$_{1-x}$O thin films using synchrotron x-ray diffraction and hard x-ray photoelectron spectroscopy
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Mg$_x$Ni$_{1-x}$O with a rock-salt structure is a wide-band-gap p-type semiconductor, performing excellent stability in the atmospheric air, good crystallinity, and transparency. Mg$_x$Ni$_{1-x}$O is a promising candidate for UV detectors, high durability hole transport layers in organic solar cells, electrochromic devices, and UV-light emitting diodes.

In this work, the effects of Mg dopant on lattice structure and electronic structures of Mg$_x$Ni$_{1-x}$O (0 ≤ x ≤ 0.52) (111) thin films were investigated. The ca. 40 nm thick films were grown epitaxially on ultra-smooth sapphire (0001) substrates by pulse laser deposition. The high-resolution x-ray reciprocal–lattice space mapping (XRSM) and hard x-ray photoelectron spectroscopy (HAXPES) were utilized. Experiments were performed at the NIMS beamline BL15XU, SPring-8 with energy resolution of 211 meV.

Upon doping with Mg, the atomic-scale structure is discussed in terms of changes in: (i) lattice distortion, (ii) crystalline mosaic spread, and (iii) static atomic disorder using Debye parameter; in addition, the electronic structure is also discussed on the basis of a Configuration-interaction model including a Zhang-Rice doublet bound state [1]. XRSM data reveals mismatch between the lattice Mg$_x$Ni$_{1-x}$O thin film and sapphire substrate is more pronounced along out-of-plane direction. The mismatch was reduced with increased Mg doping content. From the magnitude of lattice distortion we argue that the Mg$^{2+}$ ions are mainly substitute Ni$^{2+}$ ions. Heavier Mg doped films have lower Debye temperature. The lattice disorder of the films was expressed using a proposed atomic order parameter [2]. HAXPES data show that with increasing Mg content, a clear double peaks at the lowest energy of Ni 2p core-level spectra got merged. This trend is correlated to the changes observed in the valence band spectra, the peaks stand for charge transfer from band state to Ni 3d becomes closer. We assign this cumulative effect to Mg dopant in Mg$_x$Ni$_{1-x}$O, which reduce the contribution of Zhang-Rice double bound state to the Ni 3d similar to the case in Li dopant in Li$_x$Ni$_{1-x}$O [3]. However the effect of Mg dopant on charge transfer behavior is more apparent in Mg$_x$Ni$_{1-x}$O.

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