DOI: 10.1017/S1431927605504793
Microsc Microanal 11(Suppl 2), 2005
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Chemical Bonding Information from ELNES using Bond Overlap Population Diagram

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ELNES reflects the electron transition from a core-orbital to an unoccupied band. The ELNES therefore has the chemical bonding information around an objective atom. In order to reproduce and interpret the experimental ELNES, the first principles calculation of ELNES has been extensively performed. [1-13] Recently, some of theoretical studies succeeded in quantitative reproduction of the experimental spectra.[7-13] However, the relationships between spectral features and the chemical information has not been clarified in detail, even when experimental spectra are well reproduced by the calculations.

We have reported that the bond overlap population (BOP) diagram is useful for detailed analysis of the ELNES.[5,6] The BOP diagram has been computed by broadening of BOP at each energy level obtained by a first principles molecular orbital method using a cluster. For crystalline systems, calculations imposing periodic boundary condition should be a natural way. In this study, we report the BOP diagram obtained by a first principles band structure method.

First principles band structure calculation using orthogonalized linear combination of atomic orbitals (OLCAO) method is employed.[14] In this method, atomic orbitals, which are expressed by the sum of Gaussian functions, are used as basis functions. Therefore, chemical bonding information, such as BOP and net charge can be calculated in a straightforward manner. Figure 1 shows the experimental and theoretical Mg-K edge ELNES together with the BOP diagram. The calculation has been made using 128 atom-supercell, and a core-hole was introduced at a Mg-1s orbital. Since Mg-K edge ELNES reflects the electronic transition to unoccupied Mg-p orbitals, the BOP diagram between Mg-p orbital and the surrounding O-s, p and Mg-s, p, d orbitals are shown. Positive and negative values of BOP respectively corresponds to the bonding and antibonding interaction. The overlap populations among Mg* and O orbitals mainly exhibit an antibonding interaction. On the other hand, the Mg*-Mg interactions show both bonding and antibonding interactions. The shape of the wave functions in the unoccupied band is therefore well characterized by the Mg*-Mg interaction. The peaks A and D are determined by the bonding and antibonding interactions of

https://doi.org/10.1017/S1431927605504793 Published online by Cambridge University Press

Mg*p-Mg s. The peaks B and E can be mainly ascribed to the bonding and antibonding interactions of Mg*p-Mg s. The broad peak around F can be due to antibonding interactions of Mg*p-Mg d.

Origin of ELNES peaks can be interpreted in this way using BOP diagrams. BOP diagrams of AlN, GaN, InN, SiC, ZnO and their polymorphs are also discussed.

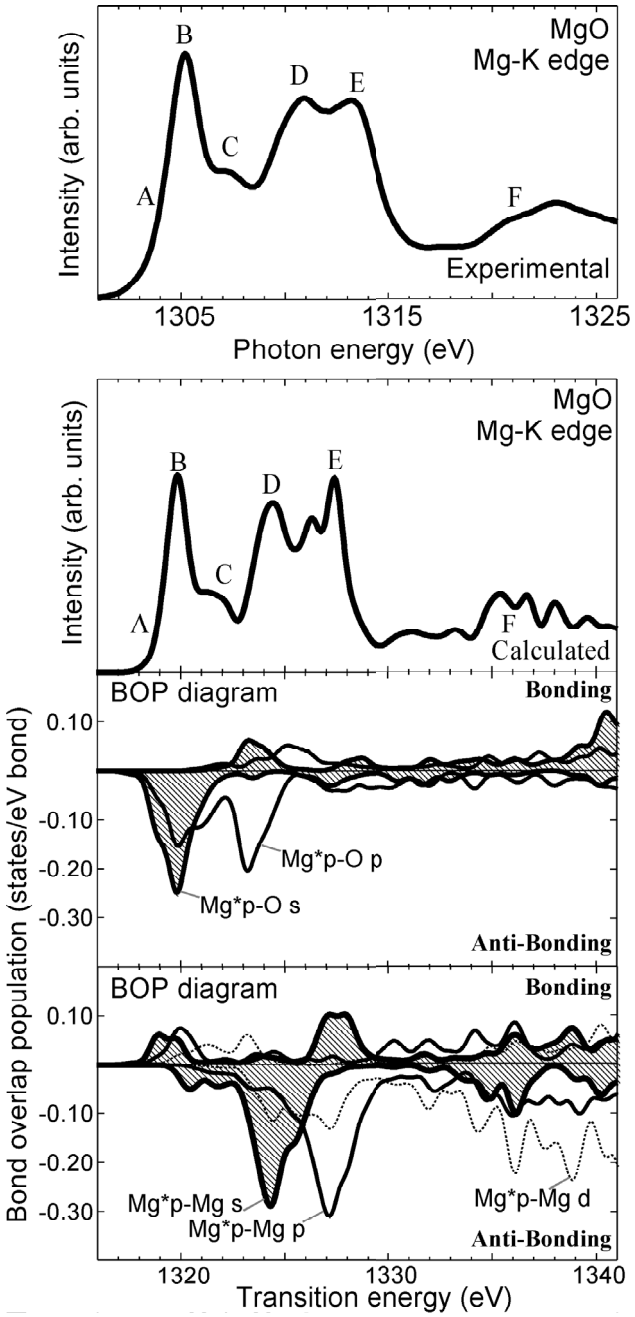

Fig. 1 (top to bottom) Experimental and theoretical Mg-K edge and the BOP diagram between Mg* and surrounding O and Mg. Mg* represents the Mg atom which includes a core-hole.

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[15] The authors thank Wai Yim Ching for allowing us to use the OLCAO program. T. M is supported as Japan Society for Promotion of Science (JSPS) fellow. This work is supported by a Grant-in-Aid for General Scientific Research from the Ministry of Education, Sports, Science and Culture of Japan.

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