Theoretical calculation and interpretation of ELNES

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Since electron-energy-loss near-edge structures (ELNES) originate from the electron transition from a core orbital to unoccupied bands, spectral features of the ELNES reflect the partial density of states of unoccupied bands, and can provide information on the atomic arrangements, electronic structures, and chemical bondings of an objective atom in materials. Recent improvements in instruments give tremendous benefits on the ELNES. The monochromator system for incident electrons achieves a high energy resolution, better than 0.3eV, and this improvement enables us to obtain much finer spectral features, namely more detailed electronic structure information. The spatial resolution of the ELNES also reaches sub-Å order with the aid of an aberration corrector for the incident electron probe. By using a modern STEM system equipped with the probe aberration corrector, ELNES observation from a single atomic column is possible. In addition to the energy and spatial resolutions, the time-resolved EELS has been developed recently. Owing to such improvements, the ELNES method is now worth to be represented as "the Ultimate Analysis", as originally proposed in 1993 by Brown.

On the other hand, to fully utilize the "ultimate analysis", a good theoretical tool is essential. Effort to reproduce and interpret the spectral profiles by theoretical calculations have been made using some theoretical frameworks [1-4]. In addition, one must know that the connection between a spectrum and information about electronic and atomic structures is not always direct or proportional because conduction band, which is responsible for ELNES, is not identical to the valence band. The relationship between the spectra and physical properties is ambiguous. The only way to find an intersection between ELNES and physical properties are detailed investigations of the electronic structures.

In this poster, one-, two-, and many-particle calculations for electron-energy-loss near-edge structures (ELNES) will be presented [4]. The most important point for the ELNES calculation is the introduction of the core-hole effect. By introducing the core-hole effect in a sufficiently large supercell, one-particle calculations are applicable to the ELNES of many edges (Fig. 1). On the other hand, the two-particle interaction between the excited electron and the core-hole, namely the excitonic effect, is significant in the K edges of very light elements and the $L_{2,3}$ edges of Mg and Al. Many-particle interactions, including both electron-electron and electron-hole interactions, are indispensable for the $L_{2,3}$ edge of transition metals and the $M_{4,5}$ edge of lanthanides, namely white lines.

Interpretation of ELNES using overlap population diagram will also be presented. The OP diagram is sometimes called "COOP (crystal orbital overlap population) diagram" [5], and a combination of band structure calculations with the OP diagram was recently proposed as a powerful method to interpret the ELNES [6-8].

References

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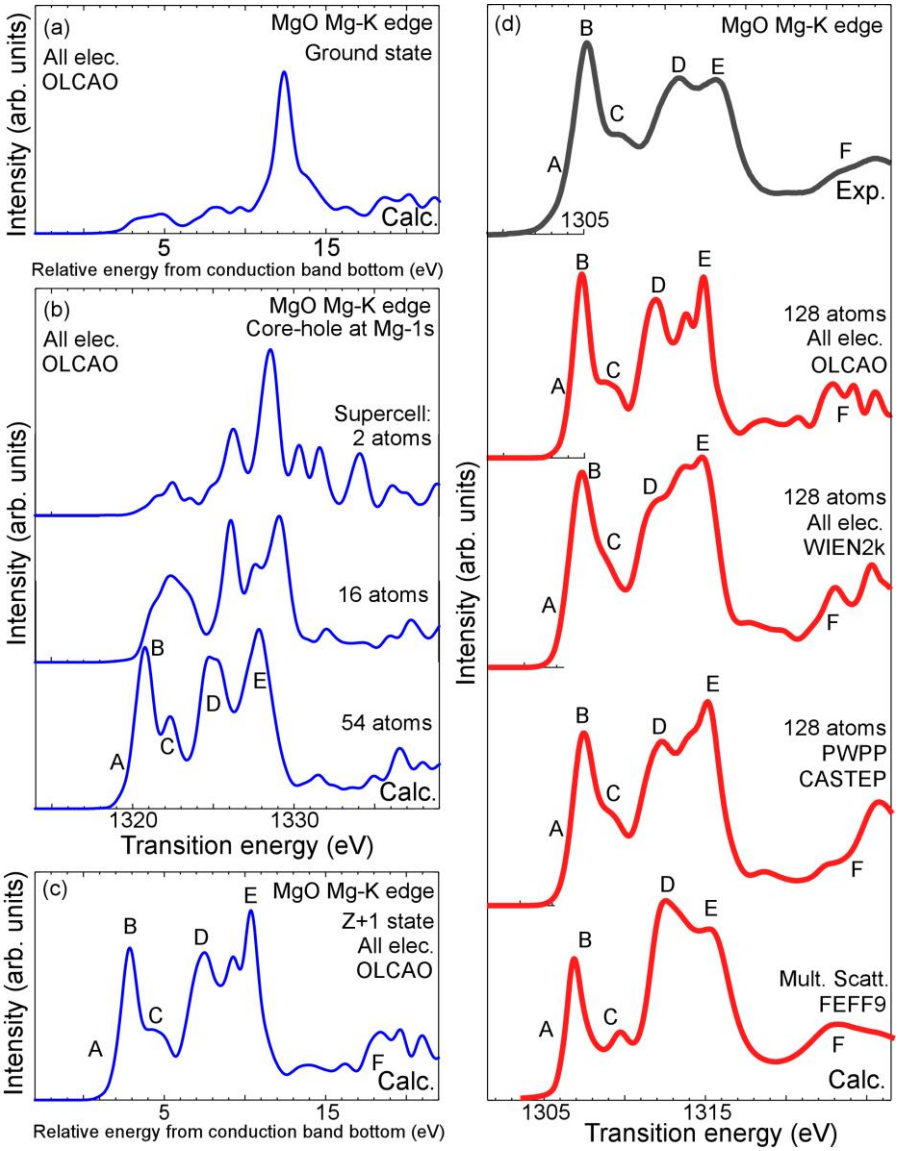

Fig.1 Calculated Mg-K edge of MgO at (a) ground state, (b) final state with different sizes of supercell, and (c) Z+1 state. (d) Experimental and calculated Mg-K edge of MgO. The calculations were performed at the final state with sufficient calculation size using the all-electron (All elec.) OLCAO method, WIEN2k code, FEFF9 code, and plane wave based pseudopotential (PWPP) method implemented in CASTEP code.