The influence of van der Waals interaction on the ELNES theoretical calculation

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The investigation of the local electronic structure of materials has been attracting attention. The electron energy-loss near edge structure (ELNES) in the electron energy-loss spectrum (EELS) reflects partial electronic state of the material and becomes one of the most powerful method to analyze the local electronic structure because ELNES is observed with a transmission electron microscopy (TEM) and scanning TEM [1], which accomplishes an atomic resolution [2]. TEM/STEM-ELNES method is possible to determine detailed electronic structures and detect their small but important changes in the interpretation of the ELNES spectra.

In order to interpret the experimental ELNES and acquire the information of electronic structures, such as the bonding state and coordination environment, a theoretical calculation of the ELNES is essential. Although, the ELNES simulation with the first-principles calculation is almost established and can reproduce the experimental spectrum with high accuracy, there is still room for improvement. In this study, we focused on the influence of van der Waals (vdW) interaction [3] on the ELNES calculation. The vdW interaction has been known to be much smaller interaction than that of ionic and covalent bonding. However, this interaction is observed in every material, and the ELNES provides a great benefit to investigate the vdW interaction at a local region.

Therefore, in this study, we investigated the effects of vdW interaction in the theoretical calculation of ELNES. Hence the vdW effect differs depending on the state, the solid, liquid and gaseous states were systematically investigated to influence of the vdW interaction on the ELNES calculation.

In the solid and liquid structures, the transition energies decreased by approximately 0.1 eV due to the influence of the vdW interaction in Fig.1(1-2). The energy shift can be interpreted as the fact that the vdW interaction exerts more influence on the excited state than the ground state. This is because the excited state has more electrons at the delocalized unoccupied bands. Furthermore, the relationships between the vdW interaction and local electronic structure and molecular configuration were discussed in Fig. 2. In contrast to the case of the solid and liquid structures, the gaseous structures are independent of the vdW interaction owing to the large intermolecular distance in Fig.1(3) [4]. We are going to talk about detailed results in my presentation.
References


FIG. 1. Experimental and calculated (1) B-K edges of h-BN in the solid state, (2) C-K edges of methanol in the liquid state and (3) C-K edges of methanol in the gaseous state. These calculated spectra were conducted with and without the vdW interaction.

FIG. 2. (1) Liquid structure of the water. The molecules are numbered as shown in the figure. (2) the relationship of the ΔTE and the Hirshfeld charge derived from the O-atom in each molecule.