Effects of interactions with surfaces on spin contamination errors and effective exchange integrals of homogeneous spin dimers, chains, and films: Model calculations of Au/MgO and Au/BaO systems

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It is well-known that unrestricted density functional theory (UDFT) calculations include a spin contamination error. While the errors in the UDFT/plane-wave calculations have not been investigated thus far, the effects of these errors are not unknown. Nevertheless, UDFT/plane-wave calculations have been applied to the theoretical investigation and design of solid materials such as electrodes and heterogeneous catalysts.

Recently, our group estimated the spin contamination errors in the UDFT/plane-wave calculation results by developing and applying of the approximate spin projection (AP) scheme [Chem. Phys. Lett., 701: 103 (2018), Mol. Phys., 117: 2251 (2019), Molecules, 24: 505 (2019), Appl. Phys. Express, 12: 115506 (2019), Chem. Lett., in press]. The AP scheme is a universal spin projection scheme for the correction of spin contamination [Chem. Lett., 15: 625 (1986), Int. J. Quant. Chem., 90: 370 (2002)]. Using the AP-UDFT/plane-wave method, the effects of spin contamination on the calculated results of heterogeneous catalysts [Chem. Phys. Lett., 701: 103 (2018), Mol. Phys., 117: 2251 (2019), Molecules, 24: 505 (2019)] and one-dimensional materials [Appl. Phys. Express, 12: 115506 (2019), Chem. Lett., in press] have been clarified. These theoretical investigations have revealed that the spin contamination errors of surface reactions are affected by the surface structures [Mol. Phys., 117: 2251 (2019)], and the interactions between the surface and adsorbates [Molecules, 24: 505 (2019)].

In this study, a more systematic theoretical investigation of the effects of the surface on the spin contamination error was performed. For this purpose, we selected model systems such as Au dimer, chain, and film adsorptions onto MgO and BaO (001) surfaces. The calculation results showed the dependence on the dimensions of the supported materials and lattice constants of the supports. The details will be presented on the day.