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Ab initio study of chemical shifts of X-ray emission spectra in ytterbium halides by the coupled cluster method

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The variety of practical applications of materials with f and heavy d elements in the composition requires the development of theoretical methods for their correct interpretation and description. However, when studying optical properties, such as excitation energies, in materials containing lanthanides and actinides, it is almost impossible to achieve reliable, highly accurate results. Currently, such calculations are only possible for molecules with few atoms due to the rapid increase in computational costs with the number of electrons explicitly considered.
For many such applications it is sufficient to simulate the electronic structure of some relatively small fragment of the crystal after performing a preliminary calculation of the periodic structure (crystal), assuming that the rest (environment of the fragment) can be considered as "frozen"with good accuracy. To apply these approaches to crystals, our laboratory has developed a technology for cutting out a fragment from a periodic structure named CTEP (compound-tunable embedding potential) method [1], which provides high accuracy in describing the influence of the environment on the selected fragment.
To analyze the correctness of reproducing the simulated electron density in the vicinity of a heavy nucleus, the chemical shift of lines of the X-ray emission spectra was chosen. For each atom, these spectra are characteristic and highly sensitive to the state of a d- or f-element in a particular compound. Direct methods for calculating the X-ray emission spectra lines are practically unapplicable, therefore a "two-step" method for its calculation was developed [2].
In this work ytterbium halides ($YbHal_n$ Hal=F, Cl, n = 2,3) are investigated by a relativistic version of the CCSD. Since this approach has limited possibilities) because of the rapid growth of its computational complexity with increasing the system size (currently, about 100 electrons can only be correlated simultaneously), therefore, it is important to carry out preliminary calculations on simpler systems and the stoichiometric molecules were chosen for this purpose. In the first part of the work, the calibration of the basic sets on molecular systems was carried out, which showed that when studying the "core-localized properties" on the Yb atom, there is no need to use saturated basic sets on halogens. In the second part of the work, pilot calculations of chemical shifts of X-ray emission spectrum lines for fragments of CTEP crystals were carried out using the relativistic CCSD.

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