Ab-initio study of Thorium activator in CaF_2 crystal an outlook for electron-nuclear bridge process.

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Thorium doped CaF_2 crystal has been recently heavily studied due to its possible application as a new time standard based on nuclear excitation of radioactive ^{229}Th isotope. Enhancing probability of excitation of nuclear isomer ^{229}Th (recently measured to be 8.35 eV [1]) requires resonance transmission of energy from electronic structure to nuclear hyperfine structure (electron bridge process). The process in theory allows for selective manipulation of hyperfine states via coupling with local electronic states induced by thorium activator in solid CaF_2 crystal. The process of doping thorium atom into the crystal matrix produces many defects which influence the absorption and emission spectra in UV and VUV spectrum. In our theoretical approach, a quantum cluster containing Th defect is being embedded into the pure CaF_2 crystal environment using ab-initio model potentials (AIMP). Electronic states are calculated utilizing multi-configurational wave function methods. In this work we discuss results of different charge compensation schemas of Thorium located in the cation site of CaF2 in the context of spectroscopic experimental data A special attention is paid to the valence of an activator located in CaF_2 lattice [2]. For the calculation we used MOLCAS software package [3], in particular the recent implementation of SCEPIC (Self-Consistent Embedded Potential for Ionic Calculation) program [4].

[1] Tiedau J. et. al. (2024) Phys. Rev. Lett. 132, 182501
[2] Veryazov V., Everestov R. A. (1991) Theor. Chim. Acta, 81, 95-103
[3] Manni G. L. et al. (2023) J. Chem. Theory Comput 19, 20, 6933-6991
[4] Larsson E. D. , Krośnicki M. , Veryazov V. (2022), A program system for self-consistent embedded potentials for ionic crystals, Chem. Phys. 562, 111549

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