Multi-configurational calculations of the electronic structure of crystalline solids; local excited states, band gaps and valency.

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Multi-configurational methods of quantum chemistry have established a significant position in ab-initio calculations of the local excited electronic states of an activator ion embedded in a crystal matrix. Barandiaran and Seijo paved the way for applying of multi-reference methods to crystalline luminescence materials by implementing ab-initio model potentials (AIMP) optimized within a self consistent calculations on perfect host [1]. The recently developed code SCEPIC [2] (Self-Consistent Embedded Potential for Ionic Calculations) along with the new MOLCAS package [3], allows for intuitive and minimal input generation of embedding potentials for ionic crystals. In this talk we will show applications of these recent developments in calculations of local defect electronic states. In particular, we will present how atomic valence redefined for crystalline solids [3] can be used to describe the properties of mixed valence compound like $Fe^{3+}[Fe^{2+}Fe^{3+}]O_4$. Moreover, we will use full valence quantity to discuss the different local environments of radioactive ^{229}Th located in CaF_2 crystal in the context of recent experiments on nuclear transitions [5].

[1] Zoila Barandiarán, Jonas Joos, Luis Seijo, Luminescent Materials: A Quantum Chemical Approach for Computer-Aided Discovery and Design, SSMATERIALS, volume 322 (2022)
[2] Larsson E. D. , Krośnicki M. , Veryazov V. (2022), A program system for self-consistent embedded potentials for ionic crystals, Chem. Phys. 562, 111549
[3] Manni G. L. et al. (2023) J. Chem. Theory Comput 19, 20, 6933-6991
[4] Veryazov V., Everestov R. A. (1991) Theor. Chim. Acta, 81, 95-103
[5] Tiedau J. et. al. (2024) Phys. Rev. Lett. 132, 182501

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