Magnetic coupling between f centers from first principles.

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The field of molecular magnetism explores more and more molecules containing lanthanide or actinide atoms. The large spin-orbit coupling and the semi-core character of the 4f and 5f orbitals confer to these molecules novel magnetic properties. Since a decade, there has been a large investigation of complexes with one magnetic center and first principles calculations have become an essential tool to interpret the experimental data, providing the nature of the ground state, the energy of exciting states and the corresponding directions of the magnetic moments.

The next experimental challenge is to gather several magnetic centers in a rational way in order to get a synergetic building of the local bricks. To achieve that, a fine knowledge of the magnetic interaction between centers is necessary. Due the semi-core character of the magnetic orbitals, the magnetic coupling between two f centers is very small, and the mechanism is still in question. The aim of this presentation is to show that quantum chemistry brings physical insights for the analysis of this magnetic coupling.

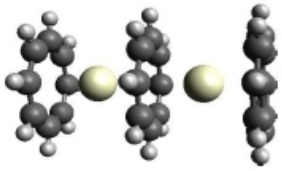

Figure 1: $[Ce_2\,COT_3]^{2-}$

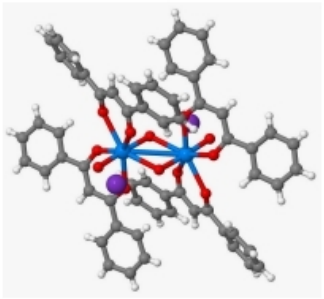

Figure 2: $[(UO_2)dbm_2 \cdot K]_2$

I will present our results on a 4f-4f dimer of Cerium(III) (Fig. 1), and a 5f-5f dimer of Uranium(V) (Fig. 2). These molecules are described using correlated wave functions based theory. The analysis of these calculations permits to determine i) the nature and magnetic properties of the local magnetic centers ii) the coupling between these centers which in each case is strongly anisotropic iii) the pathway of the magnetic coupling. In order to recover the experimental coupling, highly correlated CI methods have to be used. All calculations are simulated using Spin Hamiltonians.

References

1. F. Gendron, J. Autschbach, J. P. Malrieu, H. Bolvin *Inorg. Chem.* **58**, 581-593 (2019).