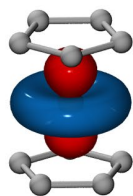
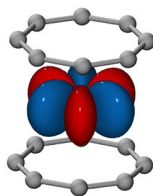


# Theory and Computation Usher In New Actinide Chemistry

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or

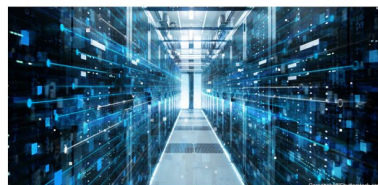


**1**  
d-Block Elements

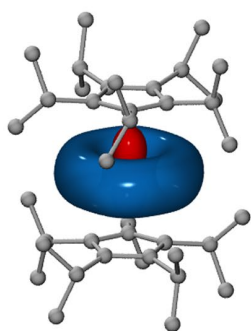
**2**  
f-Block Elements

$$\hat{H}\Psi = E\Psi$$

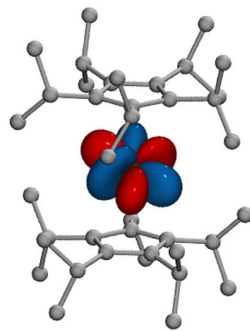
First-Principles Theory



Computation



and



**3**

**New Actinide Chemistry**

In spite of their importance for nuclear energy and radiotherapy, the chemistry of actinide elements such as uranium or plutonium has long been viewed as limited. For example, an actinide analogue of ferrocene (**1**), the quintessential organometallic compound with a *d*-valence electron configuration, has been elusive despite decades of synthetic attempts. Actinide chemistry has been thought to be constrained to mostly ionic complexes such as bis(cyclooctatetraenyl)uranium with a chemically inert *f*-valence electron configuration (**2**).

Supported by NSF Division of Chemistry, graduate student Jason M. Yu and PI Filipp Furche from the University of California, Irvine, predicted the existence of bis(pentaisopropylcyclopentadienyl) uranium (**3**), a heavy analogue of ferrocene [1]. Soon after these results became public, Layfield and co-workers reported the successful synthesis of the compound, confirming the predicted linear structure and spectroscopic properties [2]. The new uranium compound affords both 6*d* and 5*f* valence electrons and thus integrates the rich coordination chemistry of d-block elements with more traditional *f*-block element characteristics such as magnetism. This result is likely to benefit existing applications of actinides and enable new ones such as the development of catalysts for small-molecule activation or single-molecular magnets.

The ability to predict new compounds without experimental precedent from first-principles simulations is the result of long-standing efforts to develop, implement, and apply electronic structure methods by the Furche group and others. The present result arose from a combination of density functional theory backed up by advanced fluctuation-based electronic structure methods and advances in electronic structure software and algorithms enabling causal, deterministic predictions. The project also benefited from collaborations with the Long, Harvey, and Evans synthetic chemistry groups [3].

- [1] J. M. Yu, F. Furche: Theoretical Study of Divalent bis(Pentaisopropylcyclopentadienyl) Actinocenes, *Inorg. Chem.* **58**, 16004-16010, 2019.
- [2] F.-S. Guo, N. Tsoureas, G.-Z. Huang, M.-L. Tong, A. Mansikkamäki, R. A. Layfield, Isolation of a Perfectly Linear Uranium(II) Metallocene, *Angew. Chem. Int. Ed.* **59**, 2299-2303, 2020.
- [3] K. R. Gould, K. R. McClain, J. M. Yu, T. J. Groshens, F. Furche, B. G. Harvey, J. R. Long, Synthesis and Magnetism of Neutral, Linear Metallocene Complexes of Terbium(II) and Dysprosium(II), *J. Am. Chem. Soc.* **141**, 12967-12973 (2019).