**Building precise molecular architectures to engineer f-element properties**

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The complexity and richness of synthetic f-element chemistry has attracted increasing attention in recent years. This interest is stimulated jointly by scientific curiosity of a relatively unexplored field and the myriad applications that these elements have found in diverse areas such as organic synthesis, materials science and nuclear fuel cycles.1 Our research focuses on stabilising f-element complexes with unusual coordination geometries and/or oxidation states, which can provide enhanced reactivity and unique physical properties.1 We mainly utilise bulky bis(silyl)amides and cyclopentadienyls as supporting ligands to stabilise these unusual f-element motifs. Here we will present some recent highlights of this work, such as the first near-linear f-element complexes (**1**),2 the first measurements of actinide covalency by pulsed EPR spectroscopy on the Th(III) complex (**2**)3 and isolated f-block metallocenium cations (**3**), which provided record magnetic hysteresis temperatures for the dysprosium analogue in 2017.4

** (**1**) **** (**2**) (**3**)

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