

Supplementary Movie 1: Orbital Mapping of $[\text{Eu}(\text{pcam})_3\text{X}]^{2+}$ and $[\text{Eu}(\text{pcam})_3]^{3+}$. This movie reveals different energetic positions, and the shapes of unoccupied orbitals of $[\text{Eu}(\text{pcam})_3\text{X}]^{2+}$ and $[\text{Eu}(\text{pcam})_3]^{3+}$. It is created from 8000 dI/dV spectroscopic maps acquired over a pair of $[\text{Eu}(\text{pcam})_3\text{X}]^{2+}$ - $[\text{Eu}(\text{pcam})_3]^{3+}$ complexes at ± 2000 mV range with 1 mV interval between the consecutive frames.

Supplementary Movie 2: Clockwise Rotation. This movie shows controlled clockwise rotation of an $[\text{Eu}(\text{pcam})_3\text{X}_2]^+$ complex on Au(111) surface when a negative electric field is applied from the STM tip.

Supplementary Movie 3: Anticlockwise Rotation. This movie shows controlled anticlockwise rotation of an $[\text{Eu}(\text{pcam})_3\text{X}_2]^+$ complex on Au(111) surface when a negative electric field is applied from the STM tip.