

Supplementary Materials

Modelling the Effect of Zero-Field Splitting on the ^1H , ^{13}C and ^{29}Si Chemical Shifts of Lanthanide and Actinide Compounds

Table 1. EPR data for $[\text{Nd L}^{\text{Py}}]^{3+}$. Atom numbering of protons refers to Figure 1 of the main text. Hyperfine values averaged over chemically equivalent atoms.

	D/cm^{-1}	-19.830		
	(E/D)	0.127		
	g_{iso}	-0.938		
	Eigenvalues of g	-1.349	-1.290	-0.1740
$\text{H}^{(1)}$	$A_{\text{iso}}/\text{MHz}$	-0.0079		
	A_{SD}/MHz	-0.1886		
		-0.0041	0.0898	
		-0.0101	-0.0007	0.9880
$\text{H}^{(2)}$	$A_{\text{iso}}/\text{MHz}$	-0.0248		
	A_{SD}/MHz	-0.4386		
		-0.0172	0.2210	
		-0.0298	0.0020	0.2177
$\text{H}^{(3)}$	$A_{\text{iso}}/\text{MHz}$	-0.0744		
	A_{SD}/MHz	0.4201		
		0.0485	-0.2463	
		0.1290	0.0469	-0.1737
$\text{H}^{(4)}$	$A_{\text{iso}}/\text{MHz}$	-0.0190		
	A_{SD}/MHz	-0.0230		
		0.0071	0.0023	
		0.0101	-0.0060	0.0207
$\text{H}^{(5)}$	$A_{\text{iso}}/\text{MHz}$	0.0219		
	A_{SD}/MHz	-1.6432		
		-0.0743	0.8657	
		-0.1018	0.0118	0.7774
$\text{H}^{(6)}$	$A_{\text{iso}}/\text{MHz}$	-0.0274		
	A_{SD}/MHz	-0.5400		
		-0.0203	0.2800	
		-0.0325	0.0032	0.2601
$\text{H}^{(7)}$	$A_{\text{iso}}/\text{MHz}$	-0.0033		
	A_{SD}/MHz	-0.6485		
		-0.0246	0.3262	
		-0.0332	-0.0041	0.3223
$\text{H}^{(8)}$	$A_{\text{iso}}/\text{MHz}$	-0.1960		
	A_{SD}/MHz	0.3921		
		0.0191	-0.1946	
		0.0273	-0.0010	-0.1974
$\text{H}^{(9)}$	$A_{\text{iso}}/\text{MHz}$	0.0010		
	A_{SD}/MHz	2.1094		
		0.0866	-1.0539	
		0.1217	0.0043	-1.0555
$\text{H}^{(10)}$	$A_{\text{iso}}/\text{MHz}$	-0.1548		
	A_{SD}/MHz	0.4669		
		0.0224	-0.2310	
		0.0325	0.0028	-0.2360

Table 2. EPR data for tris(2,6-bis(5,6-dialkyl-1,2,4-triazin-3-yl)pyridine) uranium(III), U(MeBTP)³⁺. Atom numbering of protons refers to Figure 3 of the main text. Hyperfine values averaged over chemically equivalent atoms.

D/cm^{-1}		20.751		
(E/D)		0.219		
g^{iso}		-1.182		
Eigenvalues of g		-1.027	-1.242	-1.291
H ⁽¹⁾	$A_{\text{iso}}/\text{MHz}$	0.8704		
	A_{SD}/MHz	0.0846		
		0.0047	-0.0390	
		-0.0054	0.0018	-0.0456
H ⁽²⁾	$A_{\text{iso}}/\text{MHz}$	-0.2220		
	A_{SD}/MHz	0.2522		
		0.0062	-0.1287	
		-0.0077	-0.0027	-0.1234
H ⁽³⁾	$A_{\text{iso}}/\text{MHz}$	-0.3817		
	A_{SD}/MHz	-0.4027		
		0.0011	0.1810	
		-0.0012	-0.0113	0.2217
H ⁽⁴⁾	$A_{\text{iso}}/\text{MHz}$	-1.1959		
	A_{SD}/MHz	-0.5767		
		-0.0403	0.2391	
		0.0402	-0.0285	0.3375

Table 3. EPR data for tris(pentamethylcyclopentadienyl) uranium (III), U(C₅Me₅)₃. Hyperfine values averaged over chemically equivalent atoms.

D/cm^{-1}		289.808						
(E/D)		0.021						
g^{iso}		-0.605						
Eigenvalues of g		0.725	-1.265	-1.276				
H	$A_{\text{iso}}/\text{MHz}$	0.0454						
	A_{SD}/MHz	0.0216						
		-0.0077	-0.0429					
		0.0360	0.0165	-0.0644				
C Ring	$A_{\text{iso}}/\text{MHz}$	-0.4868						
	A_{SD}/MHz	0.1243			$A_{\text{PSO}}/\text{MHz}$	-0.4864	0.1280	-0.0726
		-0.1138	-0.3193			0.0968	-0.6695	-0.0156
		-0.0723	-0.0185	-0.4436			-0.0725	-0.0194
C Me- thyl	$A_{\text{iso}}/\text{MHz}$	0.6527						
	A_{SD}/MHz	0.0389			$A_{\text{PSO}}/\text{MHz}$	-0.0265	0.0012	-0.0002
		0.0003	0.0398			-0.0010	-0.0261	0.0009
		0.0004	-0.0004	-0.0787			-0.0007	0.0010

Table 4. EPR data for $\text{U}(\text{N}(\text{SiMe}^t\text{Bu}_2)_2)_3$. Hyperfine values averaged over chemically equivalent atoms.

D/cm^{-1}	-89.051		
(E/D)	0.212		
g_{iso}	-0.849		
Eigenvalues of g	-0.137	-0.892	-1.888
$A_{\text{iso}}/\text{MHz}$	-0.9381		
A_{SD}/MHz	-0.0477		
	0.0069	-0.1151	
	-0.1508	-0.0015	0.1630

Table 5. EPR data for Tris(trimethylsilyltetramethylcyclopentadienyl)uranium(III), $\text{U}(\text{C}_5\text{Me}_4\text{SiMe}_3)_3$. Hyperfine values averaged over chemically equivalent atoms.

D/cm^{-1}	29.714		
(E/D)	0.125		
g_{iso}	-0.739		
Eigenvalues of g	-0.272	-0.676	-1.291
$A_{\text{iso}}/\text{MHz}$	-0.5841		
A_{SD}/MHz	0.0385		
	-0.1520	0.0928	
	-0.1803	-0.0902	-0.1313

Table 6. EPR data for $\text{U}(\eta\text{-C}_5\text{Me}_4\text{Et})(\eta\text{-(1,4-C}_8\text{H}_6(\text{Si}^i\text{Pr}_3)_2)$. Hyperfine values averaged over chemically equivalent atoms.

D/cm^{-1}	-178.253		
(E/D)	0.182		
g_{iso}	-1.162		
Eigenvalues of g	-0.549	-0.769	-2.172
$A_{\text{iso}}/\text{MHz}$	-0.4951		
A_{SD}/MHz	0.0490		
	-0.1016	-0.0799	
	-0.0646	-0.1165	0.0310