Yttrium-Doped Lithium and Potassium Clusters: Magnetic Superatoms and Their Interaction with Hydrogen

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Supplementary information

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Ground state structures

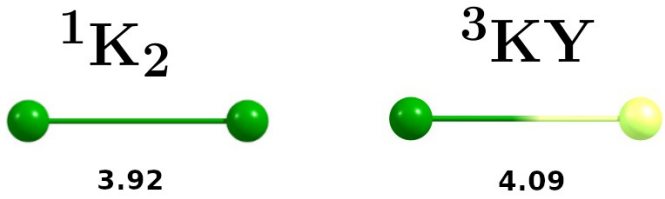

Fig. S1 K_2 and KY ground state geometries. Superscripts indicate the spin multiplicities, and selected bond lengths are given in angstroms.

Supershells

Table S1 Supershells of the clusters under study

Cluster	Supershells	Cluster	Supershells	Cluster	Supershells
K_2	$1S^2 1P^6 1D^8 2S^2$	KY	$1S^2 1P^1_\alpha 1D^1_\alpha$		
K_3	$1S^2 1P^1_\alpha$	K_2Y	$1S^2 1P^2_\alpha 1P^1_\beta$	KY_2	$1S^2 1P^4 1D^1_\alpha$
K_4	$1S^2 1P^6 1D^{10} 1F^{12} 2S^2 2P^2$	K_3Y	$1S^2 1P^2_\alpha 1P^1_\beta 1D^1_\alpha$	K_2Y_2	$1S^2 1P^5 1D^1_\alpha$
K_5	$1S^2 1P^2_\alpha 1P^1_\beta$	K_4Y	$1S^2 1P^3_\alpha 1P^2_\beta$	K_3Y_2	$1S^2 1P^6 1D^1_\alpha$
K_6	$1S^2 1P^4$	K_5Y	$1S^2 1P^6$	K_4Y_2	$1S^2 1P^6 1D^2_\alpha$
K_7	$1S^2 1P^3_\alpha 1P^2_\beta$	K_6Y	$1S^2 1P^6 1D^1_\alpha$	K_5Y_2	$1S^2 1P^6 1D^2_\alpha 1D^1_\beta$
K_8	$1S^2 1P^6$	K_7Y	$1S^2 1P^6 1D^2_\alpha$	K_6Y_2	$1S^2 1P^6 1D^3_\alpha 1D^1_\beta$
K_9	$1S^2 1P^6 1D^1_\alpha$	K_8Y	$1S^2 1P^6 1D^2_\alpha 1D^1_\beta$	K_7Y_2	$1S^2 1P^6 1D^5_\alpha$
K_{10}	$1S^2 1P^6 1D^2$	K_9Y	$1S^2 1P^6 1D^3_\alpha 1D^1_\beta$	K_8Y_2	$1S^2 1P^6 1D^5_\alpha 1D^1_\beta$
K_{11}	$1S^2 1P^6 1D^2_\alpha 1D^1_\beta$	$K_{10}Y$	$1S^2 1P^6 1D^4_\alpha 1D^1_\beta$	K_9Y_2	$1S^2 1P^6 1D^5_\alpha 1D^1_\beta 2S^1_\alpha$
K_{12}	$1S^2 1P^6 1D^4$	$K_{11}Y$	$1S^2 1P^6 1D^5_\alpha 1D^1_\beta$	$K_{10}Y_2$	$1S^2 1P^6 2S^2 1D^5_\alpha 1D^1_\beta$
K_{13}	$1S^2 1P^6 1D^3_\alpha 1D^2_\beta$	$K_{12}Y$	$1S^2 1P^6 2S^2 1D^5_\alpha$	$K_{11}Y_2$	$1S^2 1P^6 2S^2 1D^5_\alpha 1D^2_\beta$

Table S2 Global reactivity descriptors of neutral K_n ($n = 2-13$) clusters. Values are given in eV. Electronegativity (χ), hardness (η), and electrophilicity (ω) were calculated from the VIP and VEA

Cluster	χ	η	ω
K_2	2.518	1.921	1.651
K_3	2.581	1.480	2.250
K_4	2.346	1.463	1.881
K_5	2.450	1.355	2.214
K_6	2.378	1.559	1.814
K_7	2.388	1.352	2.108
K_8	2.331	1.536	2.108
K_9	2.192	1.178	2.041
K_{10}	2.267	1.202	2.137
K_{11}	2.325	1.130	2.391
K_{12}	2.358	1.230	2.262
K_{13}	2.308	1.042	2.557

Global reactivity descriptors

Table S3 Global reactivity descriptors of neutral Li_nY, K_nY ($n = 1-12$), and Li_mY_2, K_mY_2 ($m = 1-11$) clusters. Values are given in eV. Electronegativity (χ), hardness (η), and electrophilicity (ω) were calculated from the VIP and VEA

Cluster	χ	η	ω	Cluster	χ	η	ω
LiY	2.957	2.151	2.032	KY	2.663	1.838	1.930
Li_2Y	2.915	1.991	2.134	K_2Y	2.601	1.714	1.973
Li_3Y	2.910	1.990	2.127	K_3Y	2.419	1.486	1.968
Li_4Y	2.935	1.946	2.212	K_4Y	2.513	1.459	2.164
Li_5Y	2.973	1.969	2.244	K_5Y	2.475	1.394	2.196
Li_6Y	2.780	1.724	2.240	K_6Y	2.389	1.372	2.074
Li_7Y	2.813	1.670	2.368	K_7Y	2.349	1.204	2.292
Li_8Y	2.770	1.580	2.430	K_8Y	2.332	1.202	2.260
Li_9Y	2.817	1.548	2.562	K_9Y	2.318	1.136	2.365
$Li_{10}Y$	2.855	1.557	2.620	$K_{10}Y$	2.325	1.127	2.399
$Li_{11}Y$	2.853	1.557	2.615	$K_{11}Y$	2.306	1.094	2.430
$Li_{12}Y$	2.870	1.494	2.756	$K_{12}Y$	2.293	1.064	2.471
LiY_2	3.061	2.120	2.210	KY_2	2.780	1.933	1.999
Li_2Y_2	3.098	1.976	2.428	K_2Y_2	2.700	1.487	2.451
Li_3Y_2	2.875	1.796	2.301	K_3Y_2	2.572	1.466	2.257
Li_4Y_2	2.794	1.718	2.273	K_4Y_2	2.402	1.313	2.197
Li_5Y_2	2.778	1.724	2.237	K_5Y_2	2.393	1.213	2.361
Li_6Y_2	2.873	1.611	2.561	K_6Y_2	2.375	1.219	2.313
Li_7Y_2	2.815	1.631	2.428	K_7Y_2	2.318	1.153	2.330
Li_8Y_2	2.825	1.538	2.595	K_8Y_2	2.315	1.150	2.331
Li_9Y_2	2.832	1.454	2.758	K_9Y_2	2.312	1.154	2.314
$Li_{10}Y_2$	2.903	1.518	2.777	$K_{10}Y_2$	2.362	1.117	2.498
$Li_{11}Y_2$	2.976	1.471	3.009	$K_{11}Y_2$	2.383	1.095	2.592

Table S4 Atomic spin charges for the K_8Y cluster at different spin multiplicities, obtained from Mulliken and Löwdin population analyses.

Atom	El.	Spin multiplicity					
		$m_s = 2$		$m_s = 4$		$m_s = 6$	
		Mulliken	Löwdin	Mulliken	Löwdin	Mulliken	Löwdin
1	K	-0.0080	-0.0008	-0.0375	0.0321	0.4241	0.3693
2	K	0.1077	0.0765	0.3017	0.2266	0.3364	0.3160
3	K	-0.0080	0.0126	0.0809	0.1385	0.2182	0.2541
4	K	-0.0094	-0.0013	-0.0319	0.0309	0.4269	0.3701
5	K	0.1073	0.0757	0.3064	0.2288	0.3387	0.3177
6	K	0.1067	0.0759	0.3201	0.2437	0.3378	0.3179
7	K	0.1079	0.0761	0.3256	0.2466	0.3375	0.3170
8	K	-0.0102	0.0110	0.0848	0.1412	0.2255	0.2583
9	Y	0.6061	0.6743	1.6498	1.7117	2.3549	2.4796
Total		1.0001	1.0000	2.9999	3.0001	5.0000	5.0000

Table S5 Atomic spin charges for the K_7Y_2 cluster at different spin multiplicities, obtained from Mulliken and Löwdin population analyses.

Atom	El.	Spin multiplicity					
		$m_s = 2$		$m_s = 4$		$m_s = 6$	
		Mulliken	Löwdin	Mulliken	Löwdin	Mulliken	Löwdin
1	K	-0.0099	-0.0138	0.0626	0.0425	-0.0022	0.0848
2	K	0.0461	0.0505	0.2434	0.1896	0.2859	0.2241
3	K	0.0444	0.0489	0.2378	0.1864	0.2865	0.2245
4	K	0.0993	0.0278	0.0742	0.1107	0.0577	0.1577
5	K	-0.2159	-0.0446	-0.0451	0.0593	0.2911	0.2652
6	K	0.1002	0.0283	0.0718	0.1105	0.0536	0.1553
7	K	-0.2068	-0.0411	-0.0483	0.0578	0.2908	0.2652
8	Y	0.6526	0.5527	1.3442	1.2918	1.9630	2.0229
9	Y	0.4900	0.3914	1.0595	0.9514	1.7734	1.6004
Total		1.0000	1.0001	3.0001	3.0001	4.9998	5.0001

Table S6 Atomic spin charges for the $K_{10}Y$ cluster at different spin multiplicities, obtained from Mulliken and Löwdin population analyses.

Atom	El.	Spin multiplicity					
		$m_s = 2$		$m_s = 4$		$m_s = 6$	
		Mulliken	Löwdin	Mulliken	Löwdin	Mulliken	Löwdin
1	Y	0.4744	0.5425	1.5536	1.6356	2.4928	2.4427
2	K	0.0269	0.0042	-0.0917	0.0575	0.0183	0.1358
3	K	-0.0716	0.0671	-0.0868	0.0579	0.0176	0.1362
4	K	0.0927	0.0969	0.2611	0.1783	0.3210	0.3108
5	K	0.1739	0.1171	0.2664	0.1814	0.3212	0.3106
6	K	0.0294	-0.0091	0.1050	0.0937	0.3060	0.2672
7	K	0.0622	0.0181	0.1169	0.1128	0.2896	0.2549
8	K	0.0440	0.0059	0.1266	0.1046	0.3051	0.2666
9	K	0.0396	0.0068	0.0947	0.1006	0.2885	0.2543
10	K	0.0070	0.0563	0.3297	0.2400	0.3209	0.3112
11	K	0.1213	0.0942	0.3243	0.2376	0.3191	0.3098
Total		0.9998	1.0000	2.9998	3.0000	5.0001	5.0001

Table S7 Atomic spin charges for the K_9Y_2 cluster at different spin multiplicities, obtained from Mulliken and Löwdin population analyses.

		$m_s = 2$		$m_s = 4$			$m_s = 6$	
Atom	El.	Mulliken	Löwdin	El.	Mulliken	Löwdin	Mulliken	Löwdin
1	K	0.0809	0.0235	K	-0.0114	0.0774	0.2748	0.2684
2	K	-0.0229	0.0171	K	-0.0891	-0.0011	0.2558	0.3011
3	K	-0.0830	0.0215	K	0.2676	0.1922	-0.0384	0.1293
4	K	0.1243	0.0880	Y	1.3535	1.2652	0.1756	0.1771
5	K	-0.0819	0.0216	Y	1.1410	0.9517	-0.0411	0.1267
6	K	0.1290	0.0895	K	0.1921	0.1823	0.1805	0.1805
7	K	-0.1270	-0.0115	K	0.0939	0.0724	-0.0890	0.0763
8	K	-0.1206	-0.0086	K	0.1091	0.0801	-0.0922	0.0736
9	Y	0.6687	0.5132	K	-0.0486	0.0106	2.9347	2.0879
10	K	0.0905	-0.0164	K	0.0195	0.0791	0.0144	0.1808
11	Y	0.3420	0.2620	K	-0.0276	0.0901	1.4250	1.3985
Total		1.0000	1.0000		3.0000	3.0000	4.9999	5.0002

Table S8 Percentage contributions of yttrium to the density of states (DOS) of the $HOMO_\alpha$ in the K_8Y cluster. The DOS is resolved into the s, p, and d angular-momentum components of the corresponding superatomic orbital.

Multiplicity	Superorbital	Atom	% s	% p	% d
2	$HOMO_\alpha$	Y9	4.077	0.349	39.244
4		Y9	0.853	0.114	59.089
6		Y9	0.000	1.494	60.024

Table S9 Percentage contributions of yttrium to the density of states (DOS) of the $HOMO_\alpha$, together with yttrium atomic spin charges, for the K_7Y_2 cluster. The DOS is resolved into the s, p, and d angular-momentum characters of the corresponding superatomic orbital. Atomic spin charges are obtained from Mulliken (Mk) and Löwdin (Ld) population analyses.

Multiplicity	Superorbital	Atom	% s	% p	% d	μ^{Mk}	μ^{Ld}
2	$HOMO_\alpha$	Y8	0.000	1.080	41.980	0.653	0.553
2		Y9	0.000	0.484	20.721	0.490	0.391
4		Y8	0.108	1.329	33.531	1.344	1.292
4		Y9	0.483	0.875	32.492	1.059	0.951
6		Y8	0.000	2.509	33.135	1.963	2.023
6		Y9	0.000	1.563	33.052	1.773	1.600

Table S10 Percentage contributions of yttrium to the density of states (DOS) of the $HOMO_\alpha$ in the $K_{10}Y$ cluster. The DOS is resolved into the s, p, and d angular-momentum components of the corresponding superatomic orbital.

Multiplicity	Superorbital	Atom	% s	% p	% d
2	$HOMO_\alpha$	Y1	0.185	0.024	46.936
4		Y1	2.382	0.016	40.878
6		Y1	0.000	0.469	56.269

Table S11 Percentage contributions of yttrium to the density of states (DOS) of the $HOMO_\alpha$, together with yttrium atomic spin charges, for the K_9Y_2 cluster. The DOS is resolved into the s, p, and d angular-momentum characters of the corresponding superatomic orbital. Atomic spin charges are obtained from Mulliken (Mk) and Löwdin (Ld) population analyses.

Multiplicity	Superorbital	Atom	% s	% p	% d	μ^{Mk}	μ^{Ld}
2	$HOMO_\alpha$	Y9	0.970	0.475	34.520	0.6687	0.5132
2		Y11	0.141	0.707	20.782	0.3420	0.2620
4		Y4	0.001	0.600	29.904	1.3535	1.2652
4		Y5	0.016	0.190	38.260	1.1410	0.9517
6		Y9	0.000	0.112	36.442	2.9347	2.0879
6		Y11	0.000	0.040	30.959	1.4250	1.3985

Table S12 DOS contributions of yttrium and potassium to the $1P_\alpha$ and $1P_\beta$ superorbitals in K_2Y. Values are given as percentage of s, p, and d type character

Cluster	Superorbital	Atom	% s-type	% p-type	% d-type
K_2Y	$1P_\alpha$	K_1	26.943	2.470	0.550
		K_2	26.798	2.483	0.555
		Y	8.899	18.510	13.858
	$1P_\beta$	K_1	25.271	1.928	0.431
		K_2	25.205	1.944	0.434
		Y	5.345	13.590	25.853

Table S13 Superorbital energy levels of the K_9Y_2 cluster, reported in eV

α-superorbital	energy(eV)	β-superorbital	energy(eV)
$1S_\alpha$	-4.959	$1S_\beta$	-4.171
$1P_\alpha$	-3.673	$1P_\beta$	-3.230
$1P_\alpha$	-3.309	$1P_\beta$	-2.944
$1P_\alpha$	-3.249	$1P_\beta$	-2.759
$1D_\alpha$	-2.754	$1P_\beta$	-2.435
$1D_\alpha$	-2.702	$1P_\beta$	-2.166
$1D_\alpha$	-2.631		
$2S_\alpha$	-2.623		
$1D_\alpha$	-2.512		
$1D_\alpha$	-2.476		
$2D_\alpha$	-1.948		

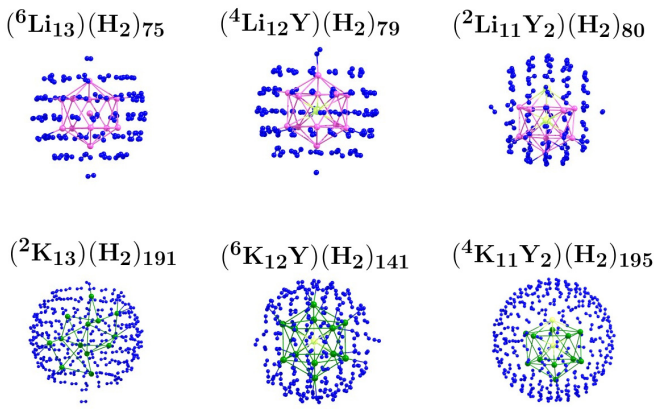

Fig. S2 Figure S2. Initial solvated configurations employed as starting points for the Born–Oppenheimer molecular dynamics simulations. In all cases, the center of mass of each cluster was first translated to the Cartesian origin. A spherical shell was then constructed around the cluster based on its maximum radial extension, and H_2 molecules were placed on this surface to form a concentric solvation environment. The number of hydrogen molecules was scaled with cluster size: 75 for $^6Li_{13}$, 79 for $^4Li_{12}Y$, 80 for $^2Li_{11}Y_2$, 191 for $^2K_{13}$, 141 for $^6K_{12}Y$, and 195 for $^4K_{11}Y_2$. The solvated structures were generated using an in-house Perl script, which randomly rotates and places H_2 molecules on the spherical surface while avoiding unphysical overlaps with the cluster atoms. These configurations were subsequently used as initial geometries for the BOMD simulations discussed in the main text.

Table S14 QTAIM criteria used to classify the cluster–H interactions in this work (all values in a.u.)

Interaction type	$\rho(r)$	$\nabla^2\rho(r)$	$\lvert V\rvert/G$	$\lvert V(r)\rvert$	$G(r)$	$H(r)$
Physisorption (closed-shell, noncovalent)	$\lesssim 0.10$	> 0.10	0.6–0.9	0.0003–0.0007	0.0004–0.0008	$+0.0002$–$+0.0003$
Chemisorption (polar / dative, partially covalent)	0.10–0.20	≥ 0	1.1–1.6	0.0010–0.0030	0.0008–0.0020	-0.0002–-0.0010
Chemisorption (open-shell, covalent)	$\gtrsim 0.20$	< 0	$\gtrsim 2.0$	0.0030–0.0080	0.0010–0.0040	-0.0020–-0.0040

Notes. (i) Signs follow standard convention: $V(r) < 0$, $G(r) > 0$, and $H(r) = V(r) + G(r)$. (ii) Ranges are representative of cluster–H bond critical points analyzed in this work. (iii) *Closed-shell* and *open-shell* describe the topological nature of the interaction rather than the electronic spin configuration. *Closed-shell* interactions ($\rho(r) \lesssim 0.10$, $\nabla^2\rho(r) > 0$, $|V|/G < 1$, $H(r) > 0$) correspond to **physisorption**, dominated by electrostatic or dispersive forces. *Open-shell* interactions ($\rho(r) \gtrsim 0.20$, $\nabla^2\rho(r) < 0$, $|V|/G > 2$, $H(r) < 0$) correspond to **chemisorption**, involving charge transfer and covalent character between the cluster and hydrogen. Intermediate values of these descriptors ($0.10 \lesssim \rho(r) \lesssim 0.20$, $1 < |V|/G < 2$) denote **polar or dative chemisorption**, a transition regime between weak and strong adsorption. (iv) Classification follows the topological and energy-density criteria proposed by R. F. W. Bader, *Chem. Rev.*, 1991, **91**, 893–928; B. Wang, X. Zhang, S. Zheng, W. Chen and M. Chen, *J. Mol. Struct.*, 2024, **1302**, 137529; and A. Kerridge, *Chem. Commun.*, 2017, **53**, 6685–6695.

Table S15 Interpretation and typical ranges of the electron localization function (ELF) according to Becke and Edgecombe

Region type	**ELF value**	**Charge behavior**	**Physical interpretation**
Core (atomic shell)	0.90–1.00	Strong charge localization	Core electrons; highly localized regions near nuclei.
Bonding region	0.70–0.90	Shared electron density	Covalent bonds; localization between bonded atoms.
Lone pairs	0.65–0.85	Localized but asymmetric charge	Non-bonding electron pairs (e.g., O, N atoms).
Delocalized / metallic region	0.40–0.65	Weak charge localization	Metallic or delocalized bonding character.
Antibonding / interstitial region	< 0.40	Charge depletion	Nodal or interstitial regions; poor localization.

Notes. (i) ELF values range from 0 (fully delocalized) to 1 (perfect localization). (ii) High ELF values correspond to regions where Pauli repulsion dominates and same-spin electrons are avoided. (iii) Visualization of ELF is complementary to QTAIM and Laplacian maps, highlighting shell structure, bonding, and lone-pair domains. (iv) Source: A. D. Becke and K. E. Edgecombe, *J. Chem. Phys.*, 1990, **92**, 5397–5403.

Table S16 Classification of chemical interactions according to the Laplacian of the electron density, $\nabla^2\rho(r)$, based on QTAIM and the criteria of Popelier and Bader

Interaction type	$\nabla^2\rho(r)$ **(a.u.)**	**Sign**	**Charge distribution**	**Character**
Covalent (shared-shell)	< 0.0	Negative	Charge concentration between nuclei	Electron sharing (bonding region).
Intermediate / dative	Slightly > 0.0	Positive	Weak charge depletion between atoms	Partially covalent / donor–acceptor.
Closed-shell (ionic, H-bond, van der Waals)	0.014–0.139	Positive	Charge depletion in the internuclear region	Electrostatic or dispersive.

Notes. (i) Negative values of $\nabla^2\rho(r)$ indicate local charge concentration and define **shared-shell (covalent)** interactions, whereas positive values correspond to **closed-shell (noncovalent)** interactions characterized by charge depletion between nuclei. (ii) The numerical ranges are adapted from P. L. A. Popelier, *J. Phys. Chem. A*, 1998, **102**, 1873–1878. (iii) Conceptual interpretation and visualization conventions follow R. F. W. Bader and C. F. Matta, *J. Phys. Chem. A*, 2004, **108**, 8385–8394. (iv) In Laplacian maps, blue regions ($\nabla^2\rho < 0$) indicate charge concentration, while red regions ($\nabla^2\rho > 0$) denote charge depletion.

Table S17 Cartesian coordinates (in Å) and CM5 atomic charges (in units of the elementary charge) for the optimized structure of the $Li_{12}Y$–$(H_2)_2$–$(H)_2$ complex. The total electronic energy and spin multiplicity are indicated above the table. Hydrogen atoms forming molecular H_2 units (H1–H4) exhibit small positive CM5 charges ($\sim +0.03\ e$), consistent with weak polarization typical of physisorbed H_2. In contrast, H5 and H6 show more negative values ($\sim -0.20\ e$), indicative of stronger interaction with the Li–Y framework. Lithium atoms display moderate positive charges, with Li_8, Li_{10}, and Li_{11} being the most polarized ($\sim +0.09$ to $+0.10\ e$), correlating with their bonding participation in the QTAIM analysis. The yttrium atom bears a large positive CM5 charge ($+27.67\ e$) due to the use of a pseudopotential including only 11 valence electrons. A physical interpretation of this value is provided in the note below

Energy: -132.29030 a.u.
Multiplicity: 2

Atom	x	y	z	CM5 charge
H1	140.180859	-141.779932	184.748113	0.02899
H2	139.633107	-142.138680	184.350227	0.02985
H3	136.995769	-139.232627	186.881526	0.02770
H4	136.292058	-139.514224	186.779139	0.02657
H5	137.335369	-140.234908	184.105457	-0.19805
H6	139.037367	-140.422969	182.074760	-0.19403
Y7	137.151800	-139.207256	182.154835	$+27.67$
Li8	139.121180	-140.287273	183.880781	0.09121
Li9	135.012294	-137.851329	180.295468	0.01785
Li10	139.881943	-139.010812	181.127079	0.09249
Li11	138.102909	-141.773594	181.146646	0.10141
Li12	134.153243	-139.155457	183.020491	0.03496
Li13	135.942367	-136.470851	182.902331	0.03493
Li14	138.959834	-137.128157	183.507296	0.03642
Li15	135.188172	-140.996306	180.665504	0.01816
Li16	136.104718	-141.525508	183.543158	0.07966
Li17	137.940923	-136.695419	180.574054	0.01174
Li18	136.660604	-138.780269	184.952438	0.04439
Li19	137.396878	-139.339536	179.080995	0.04193

Note: The large positive CM5 charge on Y ($+27.67\ e$) originates from the use of a quasirelativistic pseudopotential (ECP) that replaces 28 core electrons, leaving only 11 valence electrons explicitly included in the CM5 population analysis. Because these 28 core electrons are omitted, the apparent CM5 charge is artificially high. A physically meaningful charge can be obtained by correcting for the missing core contribution:

$$q_{\text{real}}(\text{Y}) = q_{\text{CM5}}(\text{Y}) - 28 = +27.67 - 28 = -0.33\ e.$$

Therefore, the effective charge on Y is slightly negative, indicating mild electron accumulation from the surrounding Li atoms and the adsorbed hydrogen species. All other CM5 charges (H and Li) are physically meaningful without correction, showing weakly polarized H_2 units and moderately polarized Li centers consistent with partial charge transfer within the Li–Y framework.

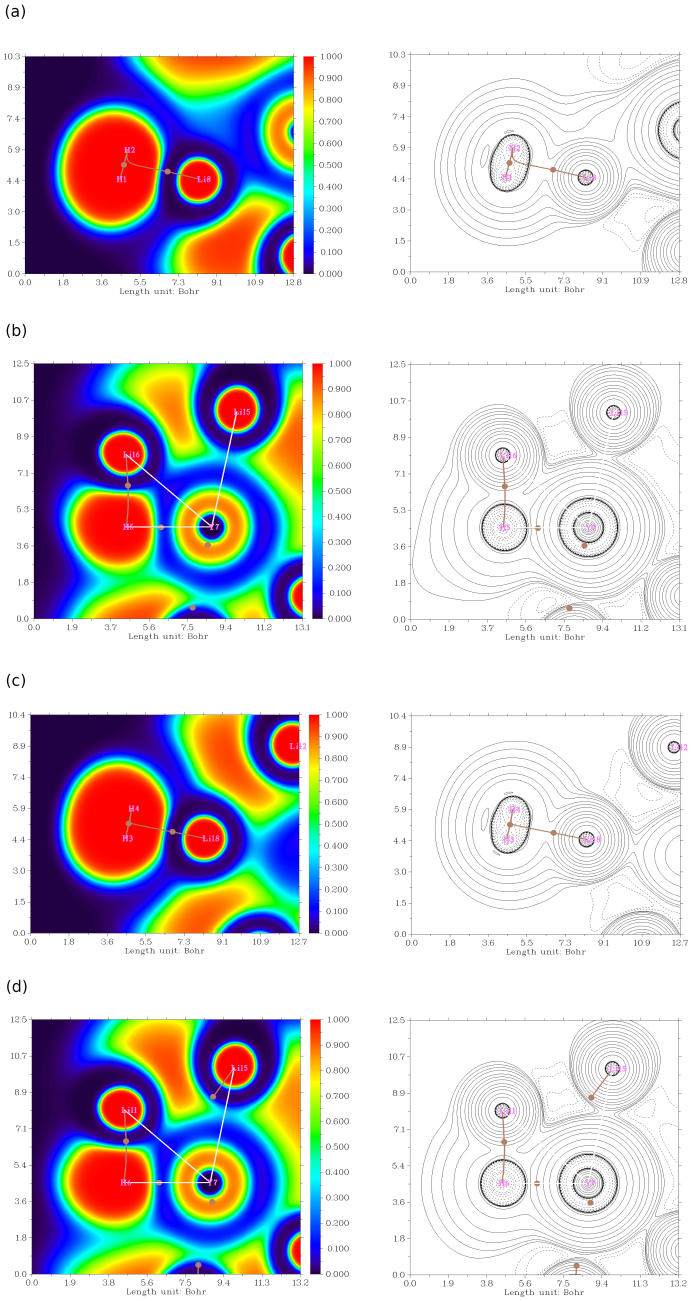

Fig. S3 Electron localization function (ELF, left) and Laplacian of the electron density, $\nabla^2\rho(r)$ (right), for selected bonding regions in the $Li_{12}Y$–$(H_2)_2$–$(H)_2$ system. (a) Molecular hydrogen H1–H2 interacts weakly with Li18, showing ELF depletion and negligible perturbation of the H–H bond, characteristic of physisorption. (b) Dissociated H5 is involved in two interactions: a polarized bond with Y7 and a weak contact with Li16, while H6 also bridges Y7 and Li11. (c) The H3–H4 dimer remains largely unperturbed, adsorbed via interaction with Li12. (d) Both H5 and H6 exhibit bonding paths to Y7, Li11, and Li15, supporting a chemisorption regime. In all cases, ELF maxima are localized on the hydrogen atoms, while depleted regions near the metal centers suggest closed-shell or polarized bonding.

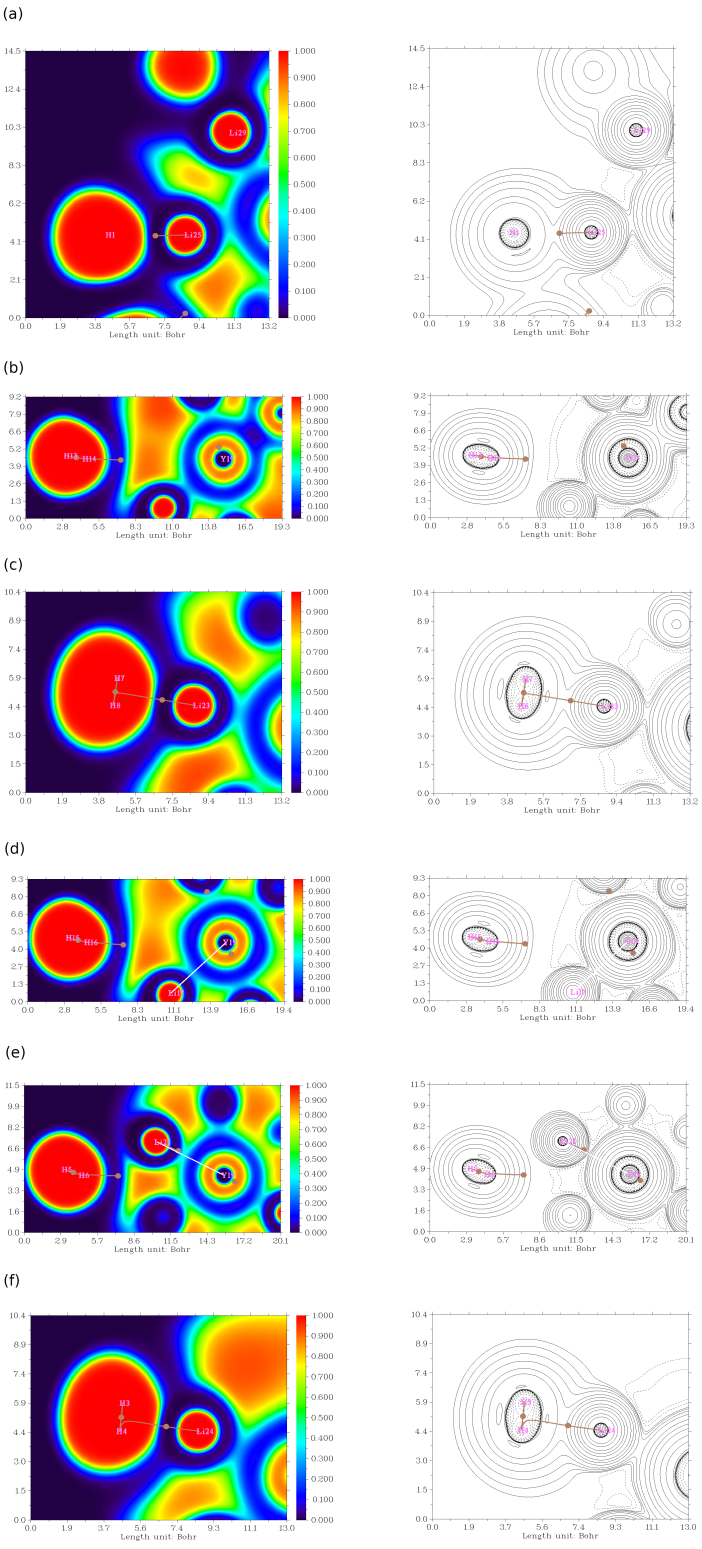

Fig. S4 Electron Localization Function (ELF, left) and Laplacian of the electron density $\nabla^2\rho(r)$ (right) maps for selected bonding regions in the $Li_{11}Y_2$–$(H_2)_8$ complex. (a) Interaction between H1 and Li25, showing a weakly polarized physisorption regime with spherical ELF lobes and low charge concentration at the BCP. (b) The Y19–H14 contact (BCP B) reveals a low ELF region and shallow charge depletion, suggesting a marginally polarized interaction with $|V|/G \approx 1$. (c) H7–H8 remains molecularly intact while interacting with Li23, exhibiting minimal ELF perturbation. (d) A diffuse ELF basin is seen between H15 and the nearby Li and Y atoms, suggesting very weak physisorption. (e) Similar features are observed for H5–H6, with faint interactions with Li21 and Y22. (f) Interaction between H3–H4 and Li24, again consistent with closed-shell, non-covalent bonding. Overall, all regions analyzed show topological and energetic characteristics of weak physisorption.

Table S18 Cartesian coordinates (in Å) and CM5 atomic charges (in units of the elementary charge) for the optimized structure of the $Li_{11}Y_2$–$(H_2)_8$ complex. The total electronic energy and spin multiplicity are given above the table. Hydrogen atoms forming molecular H_2 units (e.g., H1–H4 and H7–H12) exhibit slightly positive CM5 charges ($\sim +0.025\ e$), consistent with weak polarization and physisorption behavior. In contrast, hydrogens such as H14 and H16 display small negative values ($\sim -0.007\ e$), reflecting minimal electron accumulation near Y centers. Lithium atoms remain moderately polarized ($\sim +0.01$ to $+0.04\ e$), with Li_{17} and Li_{28} showing the highest values, consistent with their involvement in weak donor interactions identified by QTAIM analysis. Each Y atom bears a large positive CM5 charge ($+27.6$ to $+28.0\ e$), a consequence of the pseudopotential treatment with only 11 explicit valence electrons. A physical interpretation of these values is provided in the note below

Energy: −168.95053 a.u. Multiplicity: 2				
Atom	x	y	z	CM5 charge
H1	-16.084755	-15.809718	1.980865	0.02692
H2	-15.906401	-15.608420	1.273144	0.02714
H3	-12.360850	-6.726573	5.381926	0.02990
H4	-12.121262	-6.749014	4.662669	0.02673
H5	-9.981982	-11.843721	8.419168	−0.00243
H6	-10.303268	-11.894128	7.738767	−0.00648
H7	-10.680078	-13.808632	0.523136	0.02763
H8	-11.224552	-13.824644	-0.001948	0.02690
H9	-18.881115	-11.496892	1.554220	0.02410
H10	-19.304774	-11.643642	2.165351	0.02823
H11	-17.256865	-14.162163	6.951118	0.02295
H12	-16.604904	-14.530626	7.045671	0.02221
H13	-12.576179	-16.814022	1.474948	−0.00484
H14	-12.725726	-16.145430	1.795278	−0.00775
H15	-8.912902	-7.836175	4.222633	−0.00350
H16	-9.522213	-8.200596	3.965128	−0.00755
Li17	-12.370765	-9.563754	1.616926	0.03840
Li18	-14.897808	-11.339419	0.482944	0.02886
Y19	-14.227821	-11.109232	3.398469	+27.56
Li20	-15.802529	-12.949021	5.321214	0.00984
Li21	-13.358897	-11.228448	6.378190	0.03383
Y22	-15.523828	-8.603852	2.397032	+27.99
Li23	-12.333271	-12.724976	1.632080	0.00509
Li24	-13.107490	-8.649609	4.638227	0.00994
Li25	-15.095627	-13.831927	2.395704	0.01164
Li26	-15.972117	-9.849788	5.426570	0.02481
Li27	-12.919391	-13.632744	4.500852	0.00511
Li28	-11.221588	-11.139765	4.052197	0.03588
Li29	-17.161508	-11.449077	2.933737	0.01247

Note: The highly positive CM5 charges on Y_1 ($+27.56\ e$) and Y_2 ($+27.99\ e$) result from the use of a quasirelativistic pseudopotential (ECP) that replaces 28 core electrons, leaving only 11 valence electrons explicitly included in the CM5 analysis. As the core density is not represented, these apparent charges are overestimated. A physically meaningful estimate is obtained by correcting for the missing core electrons:

$$q_{\text{real}}(\text{Y}_i) = q_{\text{CM5}}(\text{Y}_i) - 28.$$

For Y_1:

$$q_{\text{real}}(\text{Y}_1) = +27.56 - 28 = -0.44\ e,$$

and for Y_2:

$$q_{\text{real}}(\text{Y}_2) = +27.99 - 28 = -0.01\ e.$$

Thus, both Y atoms are effectively neutral to slightly negative, indicating mild electron accumulation from the Li framework and the nearby H_2 molecules. All other CM5 charges (H and Li) are directly interpretable, showing weakly polarized H_2 units and moderately polarized Li centers consistent with partial charge transfer and weak physisorption.

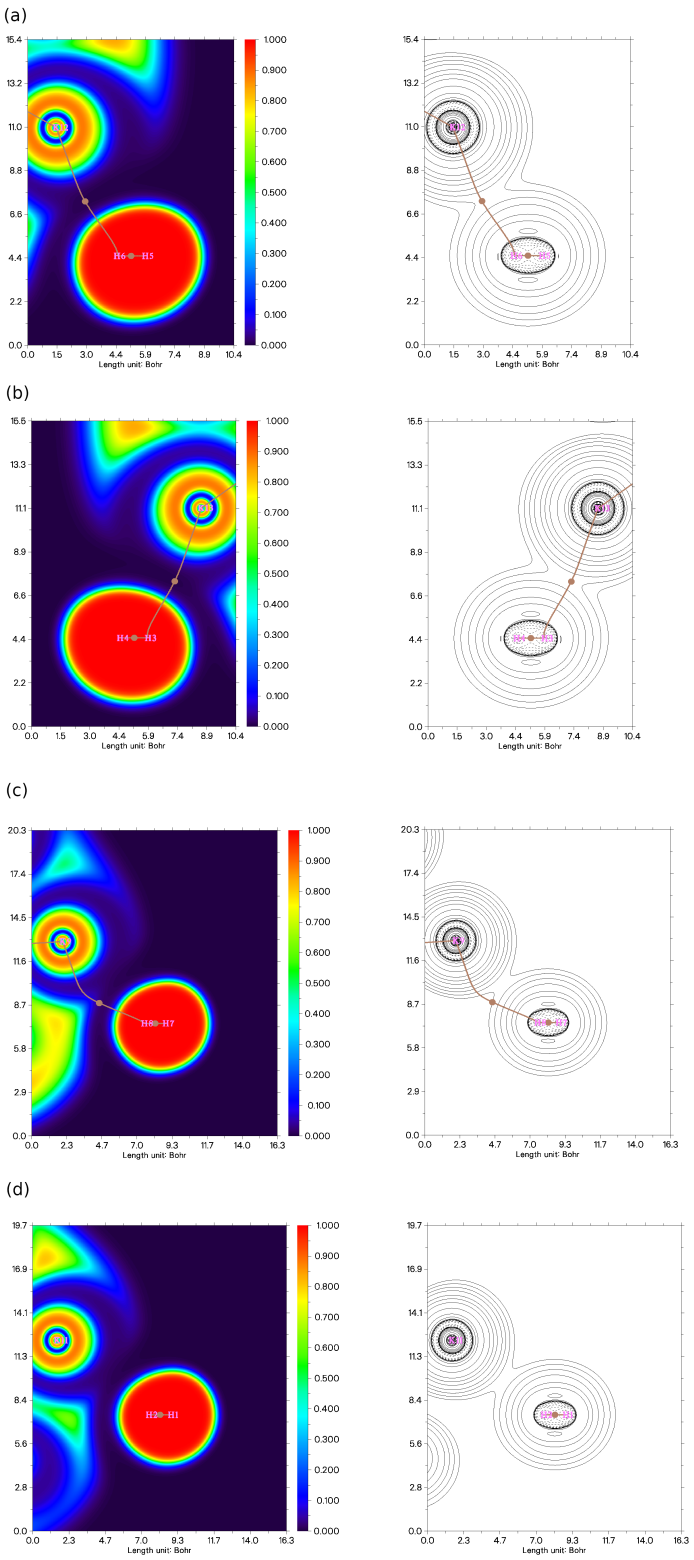

Fig. S5 Electron Localization Function (ELF, left) and Laplacian of the electron density $\nabla^2\rho(r)$ (right) maps for selected bonding regions in the $K_{12}Y–(H_2)_4$ complex. (a) K12–H6 interaction (BCP A), (b) K13–H3 interaction (BCP B), (c) K9–H8 interaction (BCP C), (d) K11–H2 interaction (BCP D). In all cases, the ELF and Laplacian contour maps show no significant electron sharing or accumulation between potassium and hydrogen atoms. The H–H bonds remain intact, with high ELF values between hydrogen atoms and charge depletion between K and H, indicating physisorption governed by weak closed-shell interactions.

Table S19 Cartesian coordinates (in Å) and CM5 atomic charges (in units of the elementary charge) for the optimized structure of the $K_{12}Y–(H_2)_4$ system. The total electronic energy and spin multiplicity are given above the table. Hydrogen atoms exhibit nearly neutral CM5 charges, consistent with weak physisorption. Potassium atoms display moderate polarization, especially K_9–K_{13}, which participate in hydrogen bonding according to QTAIM analysis. The yttrium atom bears a large positive CM5 charge (+27.61 e), originating from the use of a quasirelativistic pseudopotential with 11 explicit valence electrons

Energy: −7242.92999 a.u. Multiplicity: 4				
Atom	x	y	z	CM5 charge
H1	0.055640	0.807813	7.867708	−0.00085
H2	-0.424253	0.231622	7.935351	−0.00906
H3	-4.998524	-5.189649	-0.148382	−0.00152
H4	-4.549091	-4.869018	-0.659683	0.00373
H5	-3.999732	-6.955749	15.230968	0.00288
H6	-3.601856	-6.659823	14.664787	−0.00304
H7	-3.227530	1.286507	3.533033	−0.00127
H8	-3.131175	0.574526	3.757877	−0.00855
K9	-5.145290	-1.957062	6.228743	0.06475
K10	-1.125667	-6.463908	9.791994	0.01395
K11	-1.076197	-3.488173	6.512006	0.03466
K12	-5.244161	-6.844283	11.258646	0.05350
K13	-3.641391	-4.857687	3.352468	0.05297
K14	-1.153422	-7.628603	5.481068	0.03104
K15	-7.733040	-4.073773	9.128133	0.03046
K16	-3.741535	-9.746969	8.380655	0.05199
K17	-7.810899	-8.218495	8.097356	0.02478
K18	-7.760965	-5.237731	4.818096	0.01335
K19	-5.384045	-8.731145	4.410648	0.01429
K20	-3.504343	-2.971864	10.198197	0.01696
Y21	-4.443079	-5.850487	7.304822	+27.61

Note: The large positive CM5 charge on Y (+27.61 e) results from the use of a quasirelativistic pseudopotential (ECP) that replaces 28 core electrons, leaving only 11 valence electrons explicitly described in the CM5 population analysis. Because the core density is omitted, the apparent charge is overestimated. A physically meaningful estimate can be obtained by restoring the missing core electrons:

$$q_{\mathrm{real}}(\mathrm{Y}) = q_{\mathrm{CM5}}(\mathrm{Y}) - 28 = +27.61 - 28 = -0.39\,e.$$

Hence, the effective charge on Y is slightly negative, indicating minor electron accumulation from the surrounding K and H atoms. The other atomic charges (H and K) are directly comparable and physically meaningful, showing nearly neutral H atoms and moderately polarized K atoms.

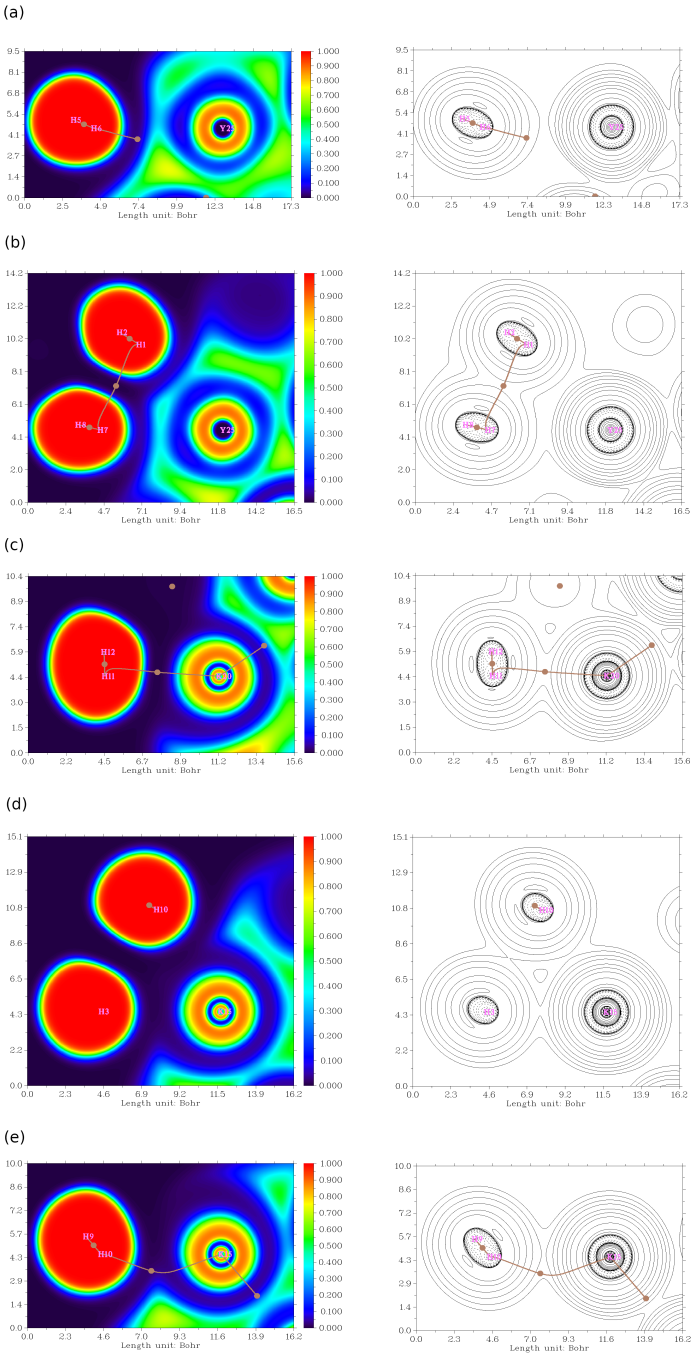

Fig. S6 Topological characterization of the $K_{11}Y_2$–$(H_2)_6$ system. Electron Localization Function (ELF) maps (left column) and contour plots of the Laplacian of the electron density, $\nabla^2\rho(r)$ (right column), for five representative interactions between the metal cluster and the adsorbed H_2 molecules: (a) Interaction with H5–H6; (b) Interaction with H1–H2 and H7–H8; (c) Interaction with H11–H12; (d) Interaction with H3–H10; (e) Interaction with H9–H10. In all cases, the ELF and Laplacian maps reveal low electronic density and charge depletion between the metal atoms and hydrogen, confirming the non-covalent, physisorptive nature of the interaction. Bond critical points (BCPs) are indicated with small spheres along the interaction paths.

Table S20 Cartesian coordinates (in Å) and CM5 atomic charges (in units of the elementary charge) for the optimized structure of the $K_{11}Y_2$–$(H_2)_6$ system. The total electronic energy and spin multiplicity are given above the table. Hydrogen atoms exhibit nearly neutral CM5 charges, consistent with weak physisorption. Potassium atoms display moderate polarization, especially those near the hydrogen molecules, as confirmed by QTAIM analysis. Each yttrium atom bears a large positive CM5 charge (+27.6 to +27.8 e), a consequence of the pseudopotential representation with 11 explicit valence electrons. A physical interpretation of these values is provided in the note below

Energy: −6683.66115 a.u. Multiplicity: 4				
Atom	x	y	z	CM5 charge
H1	-28.912903	16.225378	-37.800903	−0.01070
H2	-28.758272	16.896909	-37.486920	−0.00230
H3	-35.776066	6.937763	-36.404166	−0.00758
H4	-36.346385	6.458118	-36.295245	0.00028
H5	-28.549561	8.790604	-35.132255	−0.00307
H6	-28.957839	9.270985	-35.546346	−0.01013
H7	-30.396413	14.975518	-35.415990	−0.01069
H8	-30.326422	15.523848	-34.898983	−0.00316
H9	-38.593695	9.492833	-36.194340	0.00033
H10	-37.997129	9.919541	-36.365098	−0.00819
H11	-31.668126	18.575951	-36.412100	0.00748
H12	-31.043731	18.281367	-36.113340	0.01008
K13	-35.894224	11.812026	-42.921294	−0.02188
K14	-32.806688	8.686318	-42.565398	0.05665
K15	-35.352852	9.559653	-39.166848	0.07749
K16	-36.028839	13.893168	-39.105054	0.02801
K17	-31.946090	12.471638	-44.586541	0.02993
K18	-33.922433	15.593401	-42.408210	0.03320
K19	-29.659952	14.776836	-41.844858	0.09249
K20	-32.235983	15.680917	-38.381694	0.11521
K21	-28.965131	10.375238	-41.876324	0.07575
K22	-33.134719	11.800474	-36.325383	0.08022
K23	-31.119920	8.664231	-38.542298	0.09802
Y24	-32.363358	12.145170	-40.354910	+27.62
Y25	-29.773622	12.460027	-38.487806	+27.75

Note: The large positive CM5 charges on Y_1 (+27.62 e) and Y_2 (+27.75 e) originate from the quasirelativistic pseudopotential (ECP), which replaces 28 core electrons and leaves only 11 valence electrons explicitly included in the CM5 population analysis. Because these core electrons are not represented in the charge partitioning, the apparent values are overestimated. A physically meaningful charge can be recovered by correcting for the missing core density:

$$q_{\text{real}}(\text{Y}_i) = q_{\text{CM5}}(\text{Y}_i) - 28.$$

For Y_1:

$$q_{\text{real}}(\text{Y}_1) = +27.62 - 28 = -0.38\ e,$$

and for Y_2:

$$q_{\text{real}}(\text{Y}_2) = +27.75 - 28 = -0.25\ e.$$

Hence, both Y atoms are slightly negative, reflecting mild electron accumulation from the neighboring K atoms and adsorbed hydrogen molecules. All other CM5 charges (H and K) are physically meaningful without correction, showing nearly neutral H atoms and moderately polarized K atoms consistent with weak physisorption.

Coordinates and frequencies

Table S21 Cartesian coordinates of the Li_{13} ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-97.93943			
Multiplicity:6			
Atom	x	y	z
Li	0.000000	0.000000	0.000000
Li	0.000000	0.000000	2.893232
Li	2.577146	0.000000	1.306924
Li	-2.106887	1.501019	1.294717
Li	0.777780	2.472060	1.343032
Li	0.800527	-2.467682	1.281363
Li	-2.082309	-1.531149	1.257717
Li	2.097925	-1.510435	-1.294017
Li	-0.790995	-2.477767	-1.333692
Li	-0.008316	-0.010968	-2.896124
Li	-0.805978	2.458832	-1.281916
Li	-2.589135	-0.007946	-1.303627
Li	2.080061	1.522504	-1.253252

Table S22 Cartesian coordinates of the Li_{13} ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
1	142.1	0.1	2	143.9	0.1
3	146.4	0.1	4	150.0	0.2
5	154.2	0.5	6	169.8	0.3
7	170.6	0.1	8	173.2	0.1
9	176.9	0.0	10	178.1	0.1
11	192.1	2.9	12	192.5	9.1
13	192.9	6.4	14	194.3	25.2
15	195.9	34.3	16	199.3	4.7
17	200.3	31.5	18	203.3	0.8
19	206.2	0.1	20	208.8	0.0
21	238.3	4.7	22	243.9	2.1
23	247.1	0.2	24	249.8	1.3
25	255.1	0.8	26	269.8	0.0
27	271.4	0.0	28	272.4	0.0
29	274.9	0.0	30	275.1	0.0
31	312.3	11.1	32	314.5	13.4
33	321.2	15.1			

Table S23 Cartesian coordinates of the Li_{13} ground-state cluster optimized at the BP86/def2-TZVP-D3 level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-97.78259			
Multiplicity:6			
Atom	x	y	z
Li	-0.001296	-0.001163	0.000150
Li	-0.010726	-0.014732	2.905234
Li	2.589803	-0.000866	1.313357
Li	-2.116096	1.508087	1.299127
Li	0.782914	2.475946	1.328937
Li	0.802830	-2.478720	1.286955
Li	-2.094395	-1.543073	1.273569
Li	2.107029	-1.517309	-1.298268
Li	-0.792962	-2.481423	-1.323436
Li	0.002109	0.005329	-2.904277
Li	-0.809470	2.468715	-1.285613
Li	-2.598586	-0.007462	-1.310490
Li	2.088667	1.535139	-1.270890

Table S24 Calculated vibrational frequencies and IR intensities for the Li_{13} ground-state cluster optimized at the BP86/def2-TZVP-D3 level. Frequencies are given in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
6	149.62	0.00	7	149.98	0.00
8	150.48	0.00	9	151.04	0.00
10	151.40	0.00	11	171.16	0.00
12	171.40	0.00	13	172.15	0.00
14	172.84	0.00	15	173.19	0.00
16	185.80	0.14	17	186.21	0.00
18	186.48	0.18	19	201.91	52.14
20	202.67	51.01	21	203.26	49.97
22	207.65	0.01	23	208.37	0.01
24	208.56	0.00	25	209.11	0.00
26	243.31	0.00	27	252.96	0.00
28	254.17	0.00	29	254.97	0.00
30	256.30	0.00	31	283.73	0.00
32	284.19	0.00	33	284.31	0.00
34	284.55	0.00	35	285.08	0.00
36	327.17	6.81	37	329.69	7.12
38	331.67	7.45			

Table S25 Cartesian coordinates of the $Li_{12}Y$ ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -128.80121			
Multiplicity: 4			
Atom	x	y	z
Y	0.000000	0.000000	0.000000
Li	0.000000	0.000000	3.040451
Li	0.000000	0.018252	-3.039814
Li	-0.636805	-2.627101	1.390257
Li	-2.693089	-0.228318	1.392095
Li	0.635690	2.634453	-1.382258
Li	2.695327	0.234746	-1.386172
Li	2.272601	-1.451072	1.464296
Li	-2.272867	1.461804	-1.457016
Li	-1.105564	2.467084	1.467076
Li	1.112187	-2.459846	-1.470929
Li	2.121790	1.831141	1.248436
Li	-2.127791	-1.826362	-1.242336

Table S26 Calculated vibrational frequencies and IR intensities for the $Li_{12}Y$ ground-state cluster optimized at the PW86/DZVP level. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
1	78.9	7.7	2	81.8	11.1
3	120.9	5.0	4	121.9	7.3
5	124.3	0.9	6	126.0	0.1
7	129.9	1.1	8	132.0	3.7
9	133.9	1.7	10	141.4	1.7
11	142.2	0.8	12	143.1	0.7
13	145.2	4.6	14	150.7	4.8
15	175.8	4.2	16	184.6	3.9
17	185.1	1.3	18	203.0	17.4
19	203.9	2.1	20	210.1	19.7
21	211.5	11.5	22	231.6	18.8
23	234.0	2.9	24	268.1	0.6
25	269.2	4.1	26	273.9	2.4
27	274.1	7.4	28	275.1	14.6
29	279.7	1.3	30	281.1	0.5
31	293.1	11.6	32	296.2	4.4
33	301.8	6.9			

Table S27 Cartesian coordinates of the $Li_{12}Y$ ground-state cluster optimized at the BP86/def2-TZVP-D3 level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -128.66373			
Multiplicity: 4			
Atom	x	y	z
Y	-0.000092	0.000886	0.000230
Li	0.002045	0.000547	3.033565
Li	-0.000815	0.010956	-3.031249
Li	-0.628904	-2.621289	1.381384
Li	-2.689054	-0.224337	1.384051
Li	0.628819	2.629225	-1.373624
Li	2.689688	0.234228	-1.379855
Li	2.272156	-1.440968	1.442402
Li	-2.271925	1.449913	-1.435346
Li	-1.093648	2.465882	1.444409
Li	1.096683	-2.456798	-1.447426
Li	2.108815	1.818088	1.258369
Li	-2.112288	-1.811554	-1.252825

Table S28 Calculated vibrational frequencies and IR intensities for the $Li_{12}Y$ ground-state cluster optimized at the BP86/def2-TZVP-D3 level. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
6	105.15	7.19	7	105.87	7.24
8	113.73	0.00	9	134.11	0.00
10	137.52	0.02	11	138.79	0.07
12	139.31	0.06	13	139.73	0.28
14	140.06	1.06	15	150.07	0.12
16	150.39	0.13	17	150.80	0.00
18	151.48	0.00	19	164.23	0.00
20	195.59	0.00	21	207.17	0.00
22	217.19	0.00	23	217.40	0.00
24	220.85	4.54	25	233.93	0.01
26	235.02	0.01	27	241.36	11.59
28	241.47	11.60	29	271.50	2.16
30	275.25	0.20	31	275.37	0.28
32	275.59	0.02	33	286.07	0.00
34	289.88	0.00	35	290.46	0.00
36	303.41	18.24	37	303.77	18.43
38	307.46	27.19			

Table S29 Cartesian coordinates of the ground-state $Li_{12}Y–(H_2)_2–(H)_2$ complex optimized at the BP86/def2-TZVP-D3 level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -132.29028			
Multiplicity: 2			
Atom	x	y	z
H	140.180858	-141.779932	184.748112
H	139.633107	-142.138680	184.350226
H	136.995769	-139.232627	186.881525
H	136.292058	-139.514223	186.779138
H	137.335368	-140.234908	184.105456
H	139.037366	-140.422969	182.074759
Y	137.151799	-139.207255	182.154834
Li	139.121179	-140.287273	183.880780
Li	135.012294	-137.851329	180.295467
Li	139.881943	-139.010811	181.127078
Li	138.102908	-141.773593	181.146645
Li	134.153243	-139.155456	183.020490
Li	135.942367	-136.470850	182.902330
Li	138.959834	-137.128157	183.507295
Li	135.188171	-140.996305	180.665503
Li	136.104718	-141.525508	183.543158
Li	137.940922	-136.695418	180.574053
Li	136.660604	-138.780268	184.952437
Li	137.396878	-139.339536	179.080994

Table S30 Calculated vibrational frequencies and IR intensities for the ground-state $Li_{12}Y–(H_2)_2–(H)_2$ complex at the BP86/def2-TZVP-D3 level of theory. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
6	55.78	0.42	7	73.42	0.44
8	83.91	2.25	9	94.16	1.34
10	133.12	1.49	11	138.18	0.72
12	144.93	0.42	13	149.36	0.44
14	151.71	0.20	15	154.42	0.02
16	163.79	0.31	17	170.15	0.43
18	172.03	0.55	19	182.26	0.46
20	188.22	0.33	21	192.18	0.33
22	205.90	1.95	23	212.92	0.23
24	218.99	4.61	25	224.54	1.20
26	233.39	1.47	27	240.09	3.54
28	245.15	3.12	29	257.31	2.29
30	260.27	0.19	31	260.93	0.80
32	267.51	4.54	33	276.74	4.96
34	278.51	3.03	35	282.48	2.25
36	285.31	6.76	37	290.99	12.70
38	300.86	7.47	39	303.86	14.40
40	320.25	11.41	41	327.71	1.94
42	328.50	27.53	43	346.05	4.69
44	377.91	0.13	45	434.10	0.18
46	452.90	1.36	47	758.42	4.95
48	773.54	11.87	49	787.72	2.40
50	873.00	3.09	51	889.78	12.15
52	948.10	23.50	53	959.84	31.10
54	1010.82	2.49	55	4010.02	870.45
56	4040.59	656.98			

Table S31 Cartesian coordinates of the $Li_{11}Y_2$ ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -159.64730			
Multiplicity: 2			
Atom	x	y	z
Li	0.000000	0.000000	0.000000
Li	0.000000	0.000000	3.235070
Y	2.539055	0.000000	1.582651
Li	5.194896	-0.415642	3.060260
Li	5.132514	-0.425737	-0.065690
Y	1.189540	2.741356	1.551612
Li	0.858920	-2.563598	1.656079
Li	2.722805	1.368271	-1.050353
Li	2.585233	-1.750938	4.084068
Li	4.516275	2.236597	1.451154
Li	3.926428	-2.766727	1.532533
Li	2.385830	-1.784864	-0.948989
Li	2.963885	1.467286	4.194771

Table S32 Calculated vibrational frequencies and IR intensities for the ground-state $Li_{11}Y_2$ cluster at the PW86/DZVP level of theory. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
1	87.5	2.7	2	93.9	0.0
3	97.1	0.4	4	129.2	0.9
5	129.9	0.7	6	140.3	0.1
7	140.4	0.1	8	146.6	0.0
9	151.9	0.1	10	166.9	0.0
11	172.0	3.0	12	178.5	0.2
13	182.8	0.0	14	184.3	1.5
15	185.9	1.3	16	197.8	0.1
17	210.2	0.1	18	217.9	2.3
19	223.6	2.5	20	224.8	1.8
21	237.5	1.4	22	240.1	0.5
23	254.8	0.3	24	255.8	0.1
25	265.4	1.0	26	266.7	0.5
27	276.7	0.4	28	282.8	0.9
29	285.9	0.4	30	289.3	1.8
31	291.3	4.7	32	300.8	20.4
33	304.7	17.0			

Table S33 Cartesian coordinates of the $Li_{11}Y_2$ ground-state cluster optimized at the BP86/DEF2-TZVP-D3 level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -159.52320			
Multiplicity: 2			
Atom	x	y	z
Li	0.011728	-0.002931	-0.006375
Li	0.025774	-0.000734	3.251492
Y	2.538270	0.001177	1.577965
Li	5.179627	-0.403522	3.058443
Li	5.124957	-0.414669	-0.068266
Y	1.215777	2.688717	1.554424
Li	0.856332	-2.545687	1.656350
Li	2.720611	1.355123	-1.048936
Li	2.577155	-1.722767	4.080068
Li	4.515092	2.225554	1.451261
Li	3.917315	-2.750135	1.533279
Li	2.384907	-1.769799	-0.950242
Li	2.947835	1.445679	4.193703

Table S34 Calculated vibrational frequencies and IR intensities for the ground-state $Li_{11}Y_2$ cluster at the BP86/def2-TZVP (D3) level of theory. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
6	83.42	1.78	7	98.49	0.07
8	108.45	0.83	9	115.75	1.27
10	132.31	2.27	11	146.33	0.20
12	148.23	0.56	13	155.84	0.00
14	157.73	0.09	15	159.41	0.07
16	181.25	7.49	17	190.84	0.29
18	194.45	0.92	19	197.39	0.16
20	198.17	1.54	21	208.74	0.09
22	221.24	0.88	23	230.28	1.68
24	231.70	1.98	25	247.99	4.44
26	251.73	2.35	27	257.16	0.02
28	262.15	0.90	29	271.85	0.99
30	274.39	0.79	31	277.47	0.49
32	281.48	0.93	33	288.37	0.21
34	291.64	0.02	35	295.55	3.69
36	299.01	9.98	37	308.09	28.30
38	312.47	25.49			

Table S35 Cartesian coordinates of the $Li_{11}Y_2$–$(H_2)_8$ ground-state complex optimized at the BP86/def2-TZVP-D3 level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -168.95053			
Multiplicity: 2			
Atom	x	y	z
H	-16.084755	-15.809718	1.980865
H	-15.906401	-15.608420	1.273144
H	-12.360850	-6.726573	5.381926
H	-12.121262	-6.749014	4.662669
H	-9.981982	-11.843720	8.419168
H	-10.303267	-11.894128	7.738767
H	-10.680078	-13.808632	0.523136
H	-11.224552	-13.824644	-0.001948
H	-18.881115	-11.496892	1.554220
H	-19.304774	-11.643642	2.165351
H	-17.256865	-14.162163	6.951118
H	-16.604904	-14.530626	7.045671
H	-12.576178	-16.814022	1.474948
H	-12.725726	-16.145430	1.795278
H	-8.912902	-7.836175	4.222633
H	-9.522213	-8.200596	3.965128
Li	-12.370765	-9.563754	1.616926
Li	-14.897808	-11.339419	0.482944
Y	-14.227821	-11.109232	3.398469
Li	-15.802529	-12.949021	5.321214
Li	-13.358897	-11.228448	6.378190
Y	-15.523828	-8.603852	2.397032
Li	-12.333271	-12.724976	1.632080
Li	-13.107490	-8.649609	4.638227
Li	-15.095627	-13.831927	2.395704
Li	-15.972117	-9.849788	5.426570
Li	-12.919390	-13.632744	4.500852
Li	-11.221588	-11.139765	4.052197
Li	-17.161508	-11.449077	2.933737

Table S36 Calculated vibrational frequencies and IR intensities for the ground-state $Li_{11}Y_2$–$(H_2)_8$ complex at the BP86/def2-TZVP-D3 level of theory. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
6	4.60	0.00	7	9.46	0.08
8	18.42	0.14	9	32.44	0.11
10	34.54	0.08	11	36.96	0.41
12	44.21	0.14	13	46.78	0.05
14	48.25	0.54	15	51.74	0.17
16	54.81	0.49	17	62.88	0.44
18	65.76	1.44	19	65.94	0.53
20	77.88	2.65	21	80.66	2.16
22	82.02	2.66	23	88.72	0.54
24	90.40	0.50	25	91.58	0.96
26	93.19	0.59	27	104.46	0.09
28	105.16	0.18	29	109.74	0.12
30	123.64	1.22	31	129.36	0.86
32	132.47	0.96	33	137.84	1.11
34	138.55	0.59	35	145.17	0.18
36	149.90	0.35	37	155.55	0.09
38	156.94	0.01	39	162.38	0.26
40	164.58	0.93	41	167.00	1.25
42	174.36	2.41	43	183.75	3.74
44	187.80	0.42	45	192.38	0.94
46	193.38	0.18	47	198.99	1.15
48	201.67	1.94	49	207.06	0.59
50	211.62	0.25	51	216.02	0.54
52	227.88	0.56	53	233.38	2.49
54	243.57	3.12	55	248.01	0.51
56	249.41	7.54	57	250.61	2.32
58	257.40	0.27	59	265.55	0.81
60	270.40	1.29	61	273.22	0.84
62	274.22	2.50	63	280.39	4.09
64	282.80	3.28	65	286.13	0.65
66	288.66	2.51	67	295.50	4.19
68	299.43	4.39	69	309.87	18.61
70	315.24	4.19	71	318.75	20.39
72	325.68	34.77	73	341.97	33.65
74	372.99	9.68	75	498.62	7.37
76	511.91	8.79	77	534.08	5.54
78	583.63	5.90	79	4164.71	174.14
80	4165.90	203.71	81	4170.52	83.97
82	4188.70	78.02	83	4197.22	146.01
84	4200.49	53.28	85	4228.29	9.68
86	4232.04	92.17			

Table S37 Cartesian coordinates of the K_{13} ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-7800.74414			
Multiplicity:2			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.357258
K	4.208461	0.000000	1.980235
K	2.011159	3.702416	2.491600
K	-0.507908	3.657112	6.614716
K	-2.351590	3.717760	2.338152
K	-6.683408	2.589599	2.081130
K	-3.774279	-0.735610	2.166150
K	-4.361782	1.672260	5.897561
K	-0.043862	4.481558	-1.323883
K	-0.076235	1.213636	-4.301584
K	3.665167	2.050102	-1.854774
K	-3.853282	1.898788	-1.597702

Table S38 Calculated vibrational frequencies and IR intensities for the ground-state K_{13} cluster at the PW86/DZVP level of theory. Frequencies are given in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
1	17.2	0.0	2	17.8	0.1
3	19.7	0.1	4	23.1	0.1
5	24.0	0.1	6	26.2	0.1
7	27.5	0.5	8	28.6	0.3
9	28.7	0.2	10	30.3	0.2
11	31.8	0.1	12	32.3	0.1
13	35.5	0.1	14	35.7	0.5
15	37.1	0.1	16	37.4	0.1
17	40.7	0.0	18	41.0	0.0
19	42.1	0.0	20	44.3	0.0
21	47.7	0.0	22	48.6	0.3
23	50.8	0.0	24	53.7	0.2
25	59.2	0.0	26	60.9	0.1
27	61.7	0.0	28	63.5	0.0
29	67.2	0.1	30	68.0	0.1
31	68.6	0.1	32	74.1	0.1
33	75.3	0.2			

Table S39 Cartesian coordinates of the K_{13} ground-state cluster optimized at the BP86/def2-TZVP-D3 level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-7799.83297			
Multiplicity:2			
Atom	x	y	z
K	0.020253	0.184582	0.030766
K	-0.043755	0.047465	4.361392
K	4.005326	-0.136396	2.053851
K	2.000719	3.630652	2.438237
K	-0.442615	3.792056	6.405427
K	-2.331257	3.547402	2.350975
K	-6.599304	2.593084	2.079359
K	-3.723504	-0.705311	2.096328
K	-4.232323	1.569172	5.843675
K	-0.126021	4.462703	-1.304109
K	-0.089418	1.209197	-4.229428
K	3.604195	2.145657	-1.720330
K	-3.809854	1.907357	-1.557283

Table S40 Calculated vibrational frequencies and IR intensities for the ground-state K_{13} cluster at the BP86/def2-TZVP-D3 level of theory. Frequencies are given in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
6	21.4	0.05	7	23.3	0.04
8	24.7	0.01	9	28.1	0.15
10	29.5	0.01	11	33.8	0.13
12	35.7	0.10	13	37.1	0.05
14	37.6	0.35	15	38.8	0.07
16	42.0	0.11	17	42.1	0.17
18	43.5	0.17	19	44.8	0.02
20	45.7	0.05	21	46.9	0.09
22	48.7	0.06	23	50.2	0.00
24	51.1	0.02	25	53.5	0.02
26	56.3	0.08	27	59.1	0.03
28	59.8	0.27	29	64.6	0.08
30	71.8	0.05	31	71.9	0.12
32	72.9	0.01	33	75.5	0.01
34	77.1	0.12	35	83.2	0.11
36	84.0	0.04	37	87.9	0.31
38	89.1	0.07			

Table S41 Cartesian coordinates of the $K_{12}Y$ ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-7239.04845			
Multiplicity: 6			
Atom	x	y	z
Y	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.159079
K	3.719164	0.000000	1.866938
K	3.009819	-2.231953	-1.851276
K	-0.002150	-0.003633	-4.155076
K	-3.725317	0.002108	-1.865238
K	-3.010017	2.231669	1.858049
K	1.192072	3.783847	1.791709
K	-1.063145	3.784337	-1.873808
K	3.038284	2.376317	-1.876884
K	-1.192584	-3.778036	-1.786981
K	1.063725	-3.787515	1.878872
K	-3.038139	-2.375861	1.881058

Table S42 Calculated vibrational frequencies and IR intensities for the ground-state $K_{12}Y$ cluster at the PW86/DZVP level of theory. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
1	8.9	0.0	2	23.5	0.0
3	30.9	0.0	4	31.6	0.0
5	35.0	0.0	6	37.2	0.0
7	37.6	0.1	8	45.0	0.0
9	45.5	0.0	10	47.0	0.0
11	48.1	0.0	12	51.1	0.0
13	51.7	0.5	14	52.2	0.9
15	52.4	1.0	16	55.6	0.0
17	56.3	0.0	18	56.4	0.0
19	56.7	0.0	20	60.4	0.0
21	61.0	0.0	22	61.9	0.0
23	63.0	0.1	24	67.7	0.0
25	68.3	0.0	26	68.5	0.9
27	76.0	0.0	28	76.5	0.0
29	77.6	0.0	30	80.1	0.0
31	91.1	0.7	32	97.5	1.1
33	97.9	1.1			

Table S43 Cartesian coordinates of the $K_{12}Y$ ground-state cluster optimized at the BP86/DEF2-TZVP-D3 level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -7238.22447			
Multiplicity: 6			
Atom	x	y	z
Y	-0.001120	0.000433	0.001959
K	0.001782	0.000983	4.092437
K	3.647840	-0.000284	1.835562
K	2.953354	-2.177100	-1.819111
K	-0.003814	-0.002985	-4.088508
K	-3.651749	0.001985	-1.832656
K	-2.953363	2.176805	1.824289
K	1.168844	3.670828	1.783518
K	-1.066232	3.671347	-1.849143
K	2.987067	2.295779	-1.846783
K	-1.169638	-3.667377	-1.779201
K	1.066159	-3.673766	1.853749
K	-2.987417	-2.295367	1.850331

Table S44 Calculated vibrational frequencies and IR intensities for the ground-state $K_{12}Y$ cluster at the BP86/def2-TZVP-D3 level of theory. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
6	29.54	0.00	7	33.64	0.00
8	36.26	0.00	9	36.46	0.00
10	39.51	0.00	11	41.23	0.00
12	41.56	0.01	13	51.72	0.00
14	51.80	0.00	15	52.85	0.00
16	53.53	0.00	17	54.24	0.00
18	55.61	1.13	19	55.98	1.26
20	57.99	0.00	21	58.17	0.00
22	58.33	0.00	23	59.89	0.12
24	60.31	0.08	25	63.07	0.09
26	71.75	0.00	27	72.28	0.01
28	74.77	0.00	29	75.94	0.69
30	78.62	0.00	31	78.75	0.00
32	82.73	0.00	33	82.80	0.00
34	83.39	0.00	35	84.73	0.00
36	92.87	2.41	37	98.56	0.89
38	98.79	0.92			

Table S45 Cartesian coordinates of the $K_{12}Y–(H_2)_4$ ground-state complex optimized at the BP86/def2-TZVP-D3 level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -7242.91753			
Multiplicity: 4			
Atom	x	y	z
K	2.959900	2.522558	1.919731
K	6.121642	-0.477542	1.758175
K	1.313951	3.777821	-1.809812
K	2.079936	-1.962967	2.369342
K	3.287757	-0.222032	-1.561915
K	0.160190	-3.366885	-1.419999
K	-2.219384	-3.001335	2.026090
K	-3.746391	-1.432327	-1.597072
K	-0.383746	0.229045	-3.831489
K	-0.935978	4.245912	1.798063
K	-3.084995	2.874300	-1.759310
K	-3.984418	1.023296	1.990262
Y	-0.339880	0.449285	0.253417
H	-2.677230	-0.745226	5.849459
H	-2.458191	-0.736832	5.127894
H	-0.025147	0.813224	3.955457
H	0.024641	0.831417	4.712392
H	-6.170370	-1.287106	4.996569
H	-5.668613	-1.375418	4.441545
H	0.837133	7.799095	3.140016
H	0.644862	8.407151	3.539227

Table S46 Calculated vibrational frequencies and IR intensities for the ground-state $K_{12}Y–(H_2)_4$ complex at the BP86/def2-TZVP-D3 level of theory. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
6	6.58	0.01	34	62.40	0.22
7	9.54	0.12	35	66.61	0.29
8	11.91	0.28	36	68.89	0.06
9	17.31	0.06	37	71.79	0.32
10	19.48	0.12	38	74.04	0.14
11	21.70	0.54	39	76.48	1.45
12	25.86	0.53	40	77.20	0.02
13	28.66	0.02	41	80.82	0.57
14	31.72	0.20	42	81.51	0.24
15	34.80	2.25	43	83.09	0.87
16	38.60	0.11	44	84.87	0.16
17	40.67	0.22	45	87.72	1.26
18	41.61	0.36	46	89.98	0.52
19	43.99	0.21	47	93.26	2.94
20	44.83	0.64	48	94.98	1.28
21	45.52	0.23	49	101.48	1.09
22	46.04	1.09	50	120.94	3.34
23	48.66	1.05	51	134.94	2.78
24	49.93	0.35	52	148.88	7.87
25	52.44	0.04	53	154.05	4.52
26	53.88	1.47	54	180.73	0.99
27	55.33	0.84	55	183.96	5.01
28	55.79	0.52	56	187.80	10.24
29	56.58	1.03	57	204.34	1.51
30	58.23	0.02	58	241.18	4.38
31	59.52	0.03	59	4081.37	37.17
32	59.99	0.02	60	4211.79	36.32
33	61.61	0.19	61	4240.82	100.50
			62	4277.19	69.06

Table S47 Cartesian coordinates of the $K_{11}Y_2$ ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-6677.35043			
Multiplicity: 4			
Atom	x	y	z
K	4.593093	-0.453518	0.123560
K	2.625252	2.340768	-2.683763
K	2.411792	3.070486	1.733836
K	1.916607	-0.891354	3.700865
K	2.200416	-1.916862	-3.306658
K	1.909160	-3.950691	0.488564
K	-1.712455	-3.277509	-1.701057
K	-2.023132	-2.354623	2.786026
K	-1.218109	0.787046	-3.787306
K	-1.579783	1.894036	3.325392
K	-1.232886	3.951443	-0.433766
Y	0.062533	0.000814	0.003129
Y	-3.346418	0.315936	-0.158804

Table S48 Calculated vibrational frequencies and IR intensities for the ground-state $K_{11}Y_2$ cluster at the PW86/DZVP level of theory. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
1	28.0	0.1	2	32.4	0.0
3	34.0	0.1	4	35.8	0.3
5	36.3	0.1	6	37.9	0.1
7	40.6	0.2	8	41.2	0.3
9	42.4	0.1	10	44.2	0.4
11	46.7	0.1	12	49.6	0.0
13	51.5	0.0	14	54.1	0.1
15	54.5	0.9	16	55.8	0.1
17	59.3	0.3	18	62.3	0.2
19	64.0	0.4	20	65.6	0.1
21	65.8	0.2	22	66.9	0.2
23	69.9	0.1	24	73.7	0.1
25	74.4	0.1	26	75.2	0.4
27	75.5	0.4	28	79.4	0.2
29	80.2	0.3	30	84.6	0.3
31	100.7	2.1	32	103.2	3.0
33	121.5	0.0			

Table S49 Cartesian coordinates of the $K_{11}Y_2$ ground-state cluster optimized at the BP86/DEF2-TZVP-D3 level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-6676.59377			
Multiplicity: 4			
Atom	x	y	z
K	4.472730	-0.475642	0.100096
K	2.520006	2.310353	-2.599701
K	2.331444	3.046522	1.726412
K	1.873622	-0.807896	3.683825
K	2.185790	-1.907516	-3.253314
K	1.788423	-3.865059	0.531000
K	-1.722868	-3.162446	-1.753963
K	-1.909853	-2.421909	2.661990
K	-1.261711	0.747047	-3.731601
K	-1.558688	1.868209	3.314811
K	-1.164847	3.867106	-0.525533
Y	0.121550	0.000382	0.029267
Y	-3.069527	0.316821	-0.093272

Table S50 Modos vibracionales calculados e intensidades IR para el clúster $K_{11}Y_2$ en el nivel BP86/def2-TZVP-D3. Las frecuencias están en cm^{-1} y las intensidades en km mol^{-1}

Modo	Frecuencia (cm^{-1})	Intensidad (km mol^{-1})	Modo	Frecuencia (cm^{-1})	Intensidad (km mol^{-1})
6	32.09	0.16	7	36.92	0.00
8	37.27	0.23	9	39.02	0.01
10	39.17	0.02	11	42.28	0.25
12	42.92	0.27	13	44.56	0.10
14	47.24	0.30	15	48.45	0.00
16	52.93	0.08	17	54.18	0.18
18	55.28	0.00	19	56.16	0.54
20	56.98	0.07	21	60.29	0.43
22	63.03	0.19	23	64.64	0.68
24	68.63	0.12	25	73.85	0.64
26	74.50	0.97	27	75.48	0.09
28	77.24	0.26	29	78.38	0.24
30	79.13	0.07	31	80.50	0.26
32	83.33	0.02	33	85.17	0.06
34	85.88	0.18	35	88.52	0.50
36	99.82	2.46	37	101.16	3.67
38	133.50	0.74			

Table S51 Cartesian coordinates of the $K_{11}Y_2$–$(H_2)_6$ ground-state complex optimized at the BP86/def2-TZVP-D3 level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -6683.66115			
Multiplicity: 4			
Atom	x	y	z
H	-28.912903	16.225378	-37.800903
H	-28.758272	16.896909	-37.486919
H	-35.776066	6.937763	-36.404165
H	-36.346385	6.458118	-36.295245
H	-28.549561	8.790604	-35.132255
H	-28.957838	9.270985	-35.546346
H	-30.396413	14.975518	-35.415990
H	-30.326422	15.523848	-34.898983
H	-38.593695	9.492833	-36.194339
H	-37.997129	9.919541	-36.365098
H	-31.668126	18.575951	-36.412100
H	-31.043731	18.281367	-36.113340
K	-35.894224	11.812026	-42.921294
K	-32.806688	8.686318	-42.565398
K	-35.352852	9.559653	-39.166848
K	-36.028839	13.893168	-39.105054
K	-31.946090	12.471638	-44.586541
K	-33.922433	15.593401	-42.408210
K	-29.659952	14.776836	-41.844857
K	-32.235983	15.680917	-38.381694
K	-28.965131	10.375238	-41.876324
K	-33.134719	11.800474	-36.325382
K	-31.119920	8.664231	-38.542297
Y	-32.363358	12.145170	-40.354910
Y	-29.773622	12.460027	-38.487805

Table S52 Calculated vibrational frequencies and IR intensities for the ground-state $K_{11}Y_2$–$(H_2)_6$ complex at the BP86/def2-TZVP-D3 level of theory. Frequencies are in cm^{-1} and intensities in km mol^{-1}

Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})	Mode	Frequency (cm^{-1})	Intensity (km mol^{-1})
6	11.64	0.11	40	75.40	0.87
7	15.65	0.01	41	77.08	1.92
8	17.49	0.02	42	78.74	0.09
9	26.88	0.12	43	78.76	0.22
10	28.57	0.22	44	79.96	0.60
11	29.02	0.13	45	80.68	0.81
12	32.20	0.44	46	81.17	0.16
13	34.01	0.42	47	83.66	0.09
14	35.93	0.24	48	84.83	0.53
15	36.65	0.04	49	87.36	1.62
16	37.70	0.05	50	89.54	1.38
17	38.76	0.09	51	91.65	0.95
18	39.15	0.03	52	100.09	2.99
19	42.80	0.30	53	102.72	3.72
20	42.86	0.20	54	103.36	0.27
21	43.39	0.73	55	107.09	5.88
22	45.04	0.96	56	114.32	0.59
23	47.39	0.87	57	134.06	0.93
24	48.19	0.49	58	148.48	2.06
25	50.65	0.77	59	159.51	2.88
26	52.75	1.14	60	180.08	1.61
27	54.11	0.08	61	186.82	14.76
28	56.21	0.08	62	194.52	3.35
29	56.78	1.32	63	202.36	2.74
30	57.24	0.64	64	238.40	2.05
31	59.75	0.57	65	243.60	1.04
32	62.15	0.23	66	252.99	3.01
33	63.00	3.07	67	301.32	1.65
34	63.90	1.44	68	317.81	1.47
35	65.21	0.44	69	4135.32	123.08
36	66.19	0.90	70	4144.78	175.00
37	68.30	0.20	71	4221.05	74.69
38	74.51	0.12	72	4252.08	88.71
39	74.82	0.54	73	4252.54	62.52
			74	4275.46	1.51

Table S53 Cartesian coordinates of the K_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-1200.09922			
Multiplicity: 1			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	3.920000

Table S54 Cartesian coordinates of the K_3 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-1800.14835			
Multiplicity: 2			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.105531
K	1.635695	0.000000	-3.774317

Table S55 Cartesian coordinates of the K_4 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-2400.20666			
Multiplicity: 1			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.134118
K	3.997256	0.000000	0.734129
K	-3.998570	0.000000	3.362967

Table S56 Cartesian coordinates of the K_5 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-3000.26578			
Multiplicity: 2			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.259581
K	3.854887	0.000000	-2.131887
K	7.488295	-0.032651	0.130457
K	3.878339	-0.011991	2.430780

Table S57 Cartesian coordinates of the K_6 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-3600.32938			
Multiplicity: 1			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.245570
K	0.000000	0.424943	-4.225988
K	3.405288	1.897357	2.487049
K	3.283178	2.245991	-2.169120
K	6.434238	3.818608	0.188844

Table S58 Cartesian coordinates of the K_7 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -4200.38885			
Multiplicity: 2			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.474939
K	4.314321	0.000000	-1.183420
K	3.614765	-2.123506	2.731027
K	3.056561	2.309218	2.333430
K	2.099885	-3.768035	-1.183485
K	-0.529076	-3.793840	2.333995

Table S59 Cartesian coordinates of the K_8 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -4800.45481			
Multiplicity: 1			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.211839
K	4.197013	0.000000	-1.545129
K	3.942271	-1.292684	2.467961
K	3.764584	2.271069	5.154851
K	5.197568	3.250731	1.347489
K	1.800886	3.788494	-1.481421
K	0.592934	4.099784	2.549829

Table S60 Cartesian coordinates of the K_9 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -5400.50581			
Multiplicity: 2			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.315842
K	3.973569	0.000000	-1.835090
K	4.146544	-0.979757	2.345377
K	0.808877	-3.960067	2.613168
K	3.691604	2.449504	5.120275
K	4.988298	3.394276	1.111815
K	1.703137	3.978914	-1.646062
K	0.439708	3.900167	2.413173

Table S61 Cartesian coordinates of the K_{10} ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -6000.56506			
Multiplicity: 1			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.329407
K	4.207532	0.000000	-1.515020
K	2.805126	-4.215814	-1.229197
K	3.732175	-1.556008	2.539883
K	3.937699	1.414808	5.833446
K	2.881884	2.744056	1.761776
K	-1.447747	3.684637	2.181107
K	0.875875	4.735158	5.657992
K	0.042949	-3.934136	2.162179

Table S62 Cartesian coordinates of the K_{11} ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -6600.62489			
Multiplicity: 2			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.306932
K	4.221335	0.000000	-1.548353
K	5.598099	1.487227	2.247018
K	4.171731	-0.086178	6.044998
K	2.392993	3.748706	4.364549
K	2.376205	3.822552	-0.082008
K	3.186592	-2.332661	2.211693
K	7.356315	-2.854573	4.492413
K	7.406853	-2.734507	0.310370
K	-1.415625	3.739034	2.141170

Table S63 Cartesian coordinates of the K_{12} ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -7200.68627			
Multiplicity: 1			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.279459
K	3.801109	0.000000	-2.090950
K	2.539648	-3.619264	5.198112
K	-0.339772	-6.761008	6.216153
K	3.690767	-8.231903	6.180708
K	5.855351	-9.590996	2.789699
K	4.126456	-0.911114	2.262673
K	5.669579	-5.156278	2.406816
K	1.777857	-7.250077	2.190491
K	2.434955	-3.958988	-0.613230
K	-1.017260	-3.835780	2.220747

Table S64 Cartesian coordinates of the KY ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy:-638.33753			
Multiplicity:3			
Atom	x	y	z
Y	-0.000000	-0.000000	-0.384503
K	-0.000000	-0.000000	3.705471

Table S65 Cartesian coordinates of the K_2Y ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -1238.39873			
Multiplicity: 2			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	3.951426
Y	4.042180	0.000000	1.984323

Table S66 Cartesian coordinates of the K_3Y ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -1838.45723 Multiplicity: 3 Atom	x	y	z
K	-4.687257	-0.002717	-0.194153
K	-0.898070	-2.096787	0.165882
K	-0.893366	2.098273	0.166360
Y	2.845198	-0.000293	-0.061436

Table S67 Cartesian coordinates of the K_4Y ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -2438.52444 Multiplicity: 2 Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.118346
K	3.756119	0.000000	1.929045
K	-2.687809	-2.602883	2.128901
Y	1.502277	-3.697477	2.148207

Table S68 Cartesian coordinates of the K_5Y ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -3038.58756 Multiplicity: 1 Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.336731
K	4.391154	0.000000	0.105600
K	2.215998	3.133202	2.058403
Y	4.349857	0.210370	4.463312
K	2.348257	-2.993304	2.300929

Table S69 Cartesian coordinates of the K_6Y ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -3638.65754 Multiplicity: 2 Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.261226
K	3.977653	0.000000	-1.423227
Y	2.991899	-2.036199	2.144136
K	2.915146	2.448890	2.044695
K	4.123585	0.204593	5.575134
K	6.606581	0.147279	1.975647

Table S70 Cartesian coordinates of the K_7Y ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -4238.71847			
Multiplicity: 3			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.276858
K	3.819836	0.000000	2.134567
K	3.239581	-2.499260	-1.404729
K	5.374775	-4.093974	2.039802
K	-0.853616	-4.090363	-1.101895
K	2.435346	-6.619013	-0.025791
Y	1.278357	-3.443497	2.380020

Table S71 Cartesian coordinates of the K_8Y ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -4838.78384			
Multiplicity: 2			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.368721
K	4.154228	0.000000	-1.299719
K	1.795735	3.632474	-1.557747
K	2.913395	-2.303582	2.352074
K	3.093118	6.268256	1.703068
K	6.003710	3.970519	-0.319006
K	-0.644782	3.788112	2.024035
Y	3.128319	1.995578	2.210379

Table S72 Cartesian coordinates of the K_9Y ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -5438.84676			
Multiplicity: 3			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.269200
K	4.227471	0.000000	-1.468438
K	2.063920	-3.794785	-1.362291
Y	3.297290	-1.808610	2.163528
K	3.429407	-5.997358	2.128757
K	6.605959	-3.779969	4.104884
K	6.455973	-3.674254	-0.170155
K	2.388900	-3.560706	5.782540
K	-0.378998	-3.828340	2.236186

Table S73 Cartesian coordinates of the $K_{10}Y$ ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -6038.91190			
Multiplicity: 4			
Atom	x	y	z
Y	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.176716
K	3.683127	0.000000	1.990583
K	3.132973	2.369295	-1.685556
K	-2.977083	-2.376536	1.904600
K	-1.073524	-3.936083	-1.604317
K	1.168594	-3.762182	1.996399
K	-0.894524	3.913078	-1.681953
K	1.249619	3.767337	1.973688
K	3.047994	-2.171964	-1.799426
K	-3.007280	2.174000	1.839783

Table S74 Cartesian coordinates of the $K_{11}Y$ ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -6638.97891			
Multiplicity: 5			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.312841
K	4.366597	0.000000	-1.280541
K	2.909196	-2.377735	2.168655
K	6.867915	-0.082175	2.392320
K	6.490646	3.783225	0.088942
K	3.560133	6.205580	2.308407
K	2.328583	3.673592	5.823943
K	6.608831	4.010867	4.362607
K	2.190255	3.801369	-1.236041
K	-0.388668	3.922599	2.317466
Y	3.262337	1.912719	2.229508

Table S75 Cartesian coordinates of the KY_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -676.69333			
Multiplicity: 4			
Atom	x	y	z
K	0.000000	0.000000	0.000000
Y	0.000000	0.000000	3.959488
Y	2.947702	0.000000	2.627465

Table S76 Cartesian coordinates of the K_2Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -1276.75724			
Multiplicity: 3			
Atom	x	y	z
Y	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.054558
K	4.012510	0.000000	0.503407
Y	1.157264	2.732895	1.323739

Table S77 Cartesian coordinates of the K_3Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -1876.82809			
Multiplicity: 2			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.257146
Y	3.612800	0.000000	2.010041
Y	2.101142	-2.937533	2.006454
K	3.448144	-1.776617	5.729606

Table S78 Cartesian coordinates of the K_4Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -2476.89218			
Multiplicity: 3			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.611574
K	4.195111	0.000000	-1.113177
K	6.508523	0.002062	2.874854
Y	2.892167	1.724194	2.399029
Y	2.891893	-1.723382	2.399427

Table S79 Cartesian coordinates of the K_5Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -3076.95919			
Multiplicity: 2			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.526076
K	4.182873	0.000000	-1.327106
K	6.769835	-0.005360	2.153092
K	4.313548	0.003463	5.785821
Y	3.020172	-1.709373	2.243133
Y	3.018689	1.729649	2.250056

Table S80 Cartesian coordinates of the K_6Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -3677.02199			
Multiplicity: 3			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.278828
K	3.914169	0.000000	1.709993
K	-0.390589	4.203652	5.818725
K	2.948889	6.553649	4.495285
K	5.490037	4.190840	1.976338
Y	3.269391	2.419676	4.999130
Y	1.473597	3.358921	2.258857

Table S81 Cartesian coordinates of the K_7Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -4277.08460			
Multiplicity: 2			
Atom	x	y	z
K1	0.000000	0.000000	0.000000
K2	0.000000	0.000000	4.302083
K3	3.814651	0.000000	1.992069
K4	3.403442	-2.627483	-1.411834
K5	2.560980	-6.717456	-0.135327
K6	-2.817512	-2.636920	2.351812
K7	-1.292780	-6.718082	2.202491
Y8	1.197988	-3.434926	1.967622
Y9	-0.510712	-4.009443	-0.857235

Table S82 Cartesian coordinates of the K_7Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -4277.08470 Multiplicity: 4			
Atom	x	y	z
K1	0.000000	0.000000	0.000000
K2	0.000000	0.000000	4.372397
K3	3.791513	0.000000	2.161241
K4	3.442879	-2.693314	-1.265418
K5	2.545278	-6.879030	0.307750
K6	-2.821933	-2.669527	2.363240
K7	-1.038158	-6.864556	2.380513
Y8	1.218486	-3.423998	2.118184
Y9	-0.443333	-4.083512	-0.748042

Table S83 Cartesian coordinates of the K_7Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -4277.08569 Multiplicity: 6			
Atom	x	y	z
K1	0.000000	0.000000	0.000000
K2	0.000000	0.000000	4.496750
K3	3.937650	0.000000	2.151691
K4	3.441173	-2.558756	-1.273495
K5	3.010227	-6.465641	0.702435
K6	-2.783308	-2.529874	2.410307
K7	-0.874134	-6.442820	3.003424
Y8	1.302035	-3.193955	2.209587
Y9	-0.293459	-4.135028	-0.470040

Table S84 Cartesian coordinates of the K_8Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -4877.14995 Multiplicity: 5			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.399252
K	3.944268	0.000000	2.178934
K	3.461576	-2.564559	-1.418018
K	5.487410	-4.295324	2.135490
K	2.858441	-6.757799	0.000032
K	-2.716421	-2.578994	2.478407
K	-0.632310	-6.593960	2.853370
Y	1.411554	-3.276415	2.077461
Y	-0.341279	-4.128885	-0.590894

Table S85 Cartesian coordinates of the K_9Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -5477.21211 Multiplicity: 2 Atom	x	y	z
K1	0.000000	0.000000	0.000000
K2	0.000000	0.000000	4.230808
K3	4.256864	0.000000	-1.254220
K4	3.090197	-2.253628	2.251297
K5	1.881252	3.810370	-1.240003
K6	-0.641937	3.771869	2.257996
K7	3.211378	6.217032	2.266141
K8	6.982535	0.122956	2.271497
Y9	3.359789	2.083972	2.104625
K10	6.324304	3.920027	4.347586
Y11	5.821297	3.618208	0.230445

Table S86 Cartesian coordinates of the K_9Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -5477.21224 Multiplicity: 4 Atom	x	y	z
K1	0.000000	0.000000	0.000000
K2	0.000000	0.000000	4.218270
K3	4.257954	0.000000	-0.993085
Y4	2.827447	2.248173	2.171512
Y5	-0.319064	3.599021	2.173266
K6	1.917556	3.811340	5.760855
K7	6.058754	4.276993	4.337084
K8	6.076221	4.148448	-0.041441
K9	7.097702	0.707131	2.175524
K10	2.838614	6.353586	2.127822
K11	1.820147	3.835174	-1.460132

Table S87 Cartesian coordinates of the K_9Y_2 ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -5477.21587 Multiplicity: 6 Atom	x	y	z
K1	0.000000	0.000000	0.000000
K2	0.000000	0.000000	4.894549
K3	4.620163	0.000000	-0.656187
K4	2.635436	-2.627039	2.440443
K5	2.270789	4.038572	-0.639639
K6	-0.965604	3.600835	2.462023
K7	2.475229	5.590244	3.651770
K8	6.076622	-0.648731	3.638664
Y9	2.765002	1.608520	2.483046
K10	3.942929	2.274762	6.188844
Y11	5.571907	3.220349	2.066366

Table S88 Cartesian coordinates of the $K_{10}Y_2$ ground-state cluster optimized at the PW86/DZVP level. Energies are given in atomic units (a.u.) and coordinates in angstroms (Å)

Energy: -6077.28128			
Multiplicity: 5			
Atom	x	y	z
K	0.000000	0.000000	0.000000
K	0.000000	0.000000	4.409016
K	3.930851	0.000000	2.089996
K	-3.047379	-2.199953	2.164365
K	3.260294	-2.808348	5.610578
K	-0.959078	-4.138839	5.378933
K	3.335419	-2.505155	-1.643084
K	-0.893935	-4.276338	-1.093433
K	2.724312	-6.584848	-0.619267
K	2.100713	-6.807141	3.800394
Y	1.608557	-3.290007	1.976313
Y	4.868149	-4.163597	1.987263