Electronic Supplementary Material (ESI) for ChemComm.

Electronic Supporting Information

Predicted $M(H_2)_{12}^{n+}$ (M = Ac, Th, Pa, U, La and n = 3, 4) Complexes with Twenty Four Hydrogen Atoms Bound to the Metal Ion

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Computational Details

In the present work, all the calculations have been carried out using Turbomole 7.2 program.[1]. We have used dispersion-corrected density functional theory (DFT-D3) using BHLYP, PBE, and B3LYP with def−TZVPP basis set for optimization of all the La/Ac(III) centered H_n, (n = 1-15) clusters.[2] Relativistic effective core potential (RECP) has also been employed for heavier elements, viz., 60 core electrons for An (Ac, Th, Pa, U) and 28 core electrons for La.[3] The D3-Dispersion correction is used in all calculations because in the weakly interacted systems it can lead to significant improvements in accuracy.[4] Basis set superposition error (BSSE) has been calculated using BHLYP-D3/def-TZVPP method. For a few small systems, we have performed CCSD(T)[5], MP2[6] calculations using MOLPRO2012[7] and various DFT-D3[2] calculations. Charge calculation has been done using natural population analysis (NPA) scheme.[8] Furthermore, to obtain a clear insight into the nature of chemical bond existing between the constituent atoms quantitatively, the AIM (atoms–in–molecule) analysis[9] has been carried at BHLYP-D3/def-TZVPP level employed with energy density function (EDF)[10] utilizing Multiwfn program[11]. To investigate the nuclear quantum effect (NQE) we have used the nuclear-electronic orbital (NEO) method in conjunction with MP2 level of theory (NEO-MP2) as implemented in GAMESS-2018 software.[12] The double-zeta nuclear basis set including s, p and d functions, DZSPDN is used for the quantum hydrogen.[13] The calculation is performed using def2-TZVPP[3a] basis set for H and CRENBL basis set[14] is used for Ac with an effective core potential (ECP=78). For comparison purpose we optimized few small systems with MP2 method using CRENBL basis for Ac (ECP 78). The optimized structural parameters using CRENBL basis set (ECP 78) are found in good agreement with that of the BHLYP-D3 method (ECP 60). Moreover, to study the relativistic effect we have performed optimization using scalar relativistic ZORA approach with 4f-frozen core as well as all electron basis set using PBE-D3BJ functional and TZ2P basis set in ADF2017 software.[15] The charge calculation is also performed using Voronoi deformation density (VDD)[16] method in ADF2017.

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Table S6. Optimized Bond Lengths ($R_{(La-H),}$ $R_{min(H-H),}$ in Å), HOMO-LUMO Energy Gap (ΔE_{gap}, in eV), Charge on La (q_{La}) and Average Charge on H ($q_{avg(H)}$) Atoms (in, e) in $La(H_2)_n^{3+}$ (n = 1 - 15) Calculated using PBE-D3/Def-TZVPP Method

Table S7. Optimized Bond Lengths ($R_{(Ac-H),}$ $R_{min(H-H),}$ in Å), HOMO-LUMO Energy Gap (ΔE_{gap}, in eV), Charge on Ac (q_{Ac}) and Average Charge on H ($q_{avg(H)}$) Atoms (in, e) in $Ac(H_2)_n^{3+}$ (n = 1 - 15) Calculated using B3LYP-D3/def-TZVPP Method

Table S8. Optimized Bond Lengths ($R_{(La-H),}$ $R_{min(H-H),}$ in Å), HOMO-LUMO Energy Gap (ΔE_{gap}, in eV), Charge on La (q_{La}) and Average Charge on H ($q_{avg(H)}$) Atoms (in, e) in $La(H_2)_n^{3+}$ (n = 1 - 15) Calculated using B3LYP-D3/def-TZVPP Method

Table S9. Calculated Value of Relative Energy (RE, in eV) of Mixed Species ($[Ac(H)_2(H_2)_y^{3+}]$ and $[Ac(H)_4(H_2)_y^{3+}]$, where y = 1, 2, 9-10) with Respect to the Corresponding Similar Composition $Ac(H_2)_n^{3+}$ System and their Binding Energy (BE, in eV) at BHLYP/def-TZVPP Level of Theory. Binding Energy of $Ac(H_2)_n^{3+}$ (BE, in eV) Systems is Provided within Parenthesis

Table S10. Calculated Value of Optimized Bond Lengths (in Å), HOMO-LUMO Gap (ΔE_{gap}, in eV), Charge on Metal ion (q_{Ac}, in e) and Average NPA Charge on H atom (q_H, in e) and on H atom of H_2 Molecule (q_{H2}, in e) in Mixed Species at BHLYP-D3/def-TZVPP Level of Theory

Table S11. Binding Energy and Binding Energy per H_2 Molecule (BE, in eV) of $Ac(H_2)_n^{3+}$ (n = 1 - 12) Calculated using PBE-D3, B3LYP-D3, BHLYP-D3 Functionals with def-TZVPP Basis Set.

Table S12. Binding Energy and Binding Energy per H_2 Molecule (BE, in eV) of $La(H_2)_n^{3+}$ (n = 1 - 12) Calculated using PBE-D3, B3LYP-D3, BHLYP-D3 Functionals with def-TZVPP Basis Set.

Table S13. Basis Set Superimposition Error (BSSE, in eV) Calculated Using BHLYP-D3/def-TZVPP Method.

Table S14. Binding Energy (BE, in eV) and Binding Energy per H_2 Molecule (BE/H_2, in eV) Calculated Using MP2 and NEO-MP2 Method with def2-TZVPP Basis set for H and CRENBL Basis set of Ac. (DZSPDN Nuclear Basis set is used for the Quantum Hydrogen)

Table S15. Calculated Values of Bond Critical Point Electron Density (ρ in e a_0^{-3}), Laplacian of Electron Density ($\nabla^2\rho$ in e a_0^{-5}), Local Electron Energy Density (E_d in au), and Ratio of Local Electron Kinetic Energy Density and Electron Density ($G(r)/\rho$ in au) of $Ac(H_2)_n^{3+}$ (n = 1 - 12), $Th(H_2)_{12}^{3+}$, $Th(H_2)_{12}^{4+}$, $Pa(H_2)_{12}^{4+}$, $U(H_2)_{12}^{4+}$ Clusters as obtained by using BHLYP-D3/def-TZVPP Method along with Small Core ECP Employed with EDF

Table S16. Calculated Values of Bond Critical Point Electron Density (ρ in e a_0^{-3}), Laplacian of Electron Density ($\nabla^2\rho$ in e a_0^{-5}), Local Electron Energy Density (E_d in au), and Ratio of Local Electron Kinetic Energy Density and Electron Density ($G(r)/\rho$ in au) of $La(H_2)_n^{3+}$ (n = 1 - 12) Clusters as obtained by using BHLYP-D3/def-TZVPP Method along with Small Core ECP Employed with EDF

Table S17. Calculated Value of Optimized Bond Lengths (in Å), Binding Energy (BE, in eV), Binding Energy Per Hydrogen Molecule (BE/H_2, in eV), Energy Gain on H_2 addition (EG, in eV), HOMO-LUMO Energy Gap ($\Delta E_{gap,}$ in eV) and Voronoi Deformation Density Charge (VDD) on Metal ion (q_M, in e) in $Ac(H_2)_n^{3+}$ (n = 1-12) and $M(H_2)_{12}^{3+/4+}$ (M = La, Th, U) System using Scalar Relativistic ZORA Approach at PBE-D3BJ/TZ2P at Level of Theory with 4f-Frozen Core

Table S18. Calculated Value of Optimized Bond Lengths (in Å), Binding Energy (BE, in eV), Binding Energy Per Hydrogen Molecule (BE/H_2, in eV), Energy Gain on H_2 addition (EG, in eV), HOMO-LUMO Energy Gap ($\Delta E_{gap,}$ in eV) and Voronoi Deformation Density Charge (VDD) on Metal ion (q_M, in e) in $Ac(H_2)_n^{3+}$ (n = 1-12) and $M(H_2)_{12}^{3+/4+}$ (M = La, Th, U) System using Scalar Relativistic ZORA Approach with All Electron Basis Set at PBE-D3BJ/TZ2P Level of Theory

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Table S1. Optimized Bond Lengths ($R_{(Ac-H)}$, $R_{min(H-H)}$, in Å), Binding Energy (BE, in eV), of $Ac(H_2)_n^{3+}$ (n = 1-3) Calculated using MP2, CCSD(T) Methods and PBE-D3, PBE0-D3, B3LYP-D3, BHLYP-D3, TPSS-D3, TPSSH-D3 Functional with def-TZVPP Basis Set.

Methods	$R_{min(Ac-H)}$	$R_{max(Ac-H)}$	$R_{min(H-H)}$	BE (eV)
$Ac(H_2)^{3+}$				
PBE-D3	2.716	2.716	0.786	-0.939
B3LYP-D3	2.734	2.734	0.774	-0.845
TPSS-D3	2.701	2.701	0.776	-0.895
PBE0-D3	2.700	2.700	0.778	-0.887
TPSSH-D3	2.698	2.698	0.774	-0.878
BHLYP-D3	2.722	2.722	0.766	-0.809
MP2	2.722	2.722	0.766	-0.776
CCSD(T)	2.724	2.724	0.771	-0.780
$Ac(H_2)_2^{3+}$				
PBE-D3	2.726	2.730	0.783	-1.805
B3LYP-D3	2.761	2.764	0.772	-1.628
TPSS-D3	2.704	2.719	0.774	-1.720
PBE0-D3	2.708	2.715	0.776	-1.709
TPSSH-D3	2.701	2.717	0.772	-1.689
BHLYP-D3	2.753	2.755	0.764	-1.563
MP2	2.727	2.733	0.764	-1.512
CCSD(T)	2.729	2.736	0.770	-1.520
$Ac(H_2)_3^{3+}$				
PBE-D3	2.727	2.743	0.781	-2.610
B3LYP-D3	2.746	2.760	0.770	-2.354
TPSS-D3	2.711	2.724	0.772	-2.489
PBE0-D3	2.715	2.727	0.774	-2.477
TPSSH-D3	2.714	2.726	0.770	-2.446
BHLYP-D3	2.735	2.746	0.762	-2.270
MP2	2.737	2.744	0.763	-2.211
CCSD(T)	2.739	2.746	0.768	-2.223

Table S2. Optimized Bond Lengths ($R_{(Ac-H)}$, $R_{min(H-H)}$, in Å), HOMO-LUMO Energy Gap (ΔE_{gap}, in eV), Charge on Ac (q_{Ac}) and Average Charge on H ($q_{avg(H)}$) Atoms (in, e) in $Ac(H_2)_n^{3+}$ (n = 1 - 12) Calculated using BHLYP-D3/def-TZVPP Method

System	$R_{(Ac-H)}$		$R_{min(H-H)}$	ΔE_{gap}	q_{La}	$q_{avg(H)}$
	R_{min}	R_{max}				
$Ac@(H_2)^{3+}$	2.722	2.722	0.766	11.600	2.963	0.019
$Ac@(H_2)_2^{3+}$	2.753	2.755	0.764	11.559	2.916	0.021
$Ac@(H_2)_3^{3+}$	2.735	2.746	0.762	11.761	2.852	0.025
$Ac@(H_2)_4^{3+}$	2.741	2.759	0.760	11.822	2.785	0.027
$Ac@(H_2)_5^{3+}$	2.742	2.780	0.758	11.847	2.721	0.028
$Ac@(H_2)_6^{3+}$	2.763	2.781	0.757	12.014	2.631	0.031
$Ac@(H_2)_7^{3+}$	2.765	2.789	0.756	12.163	2.539	0.033
$Ac@(H_2)_8^{3+}$	2.777	2.802	0.755	12.129	2.440	0.035
$Ac@(H_2)_9^{3+}$	2.787	2.806	0.754	12.567	2.325	0.038
$Ac@(H_2)_{10}^{3+}$	2.788	2.819	0.753	12.205	2.205	0.040
$Ac@(H_2)_{11}^{3+}$	2.790	2.837	0.751	12.166	2.077	0.042
$Ac@(H_2)_{12}^{3+}$	2.815	2.828	0.750	12.977	1.932	0.044

Table S3. Optimized Bond Lengths ($R_{(La-H)}$, $R_{min(H-H)}$, in Å), HOMO-LUMO Energy Gap (ΔE_{gap}, in eV), Charge on La (q_{La}) and Average Charge on H ($q_{avg(H)}$) Atoms (in, e) in $La(H_2)_n^{3+}$ (n = 1 - 15) Calculated using BHLYP-D3/def-TZVPP Method

System	$R_{(La-H)}$		$R_{min(H-H)}$	ΔE_{gap}	q_{La}	$q_{avg(H)}$
	R_{min}	R_{max}				
$La(H_2)^{3+}$	2.627	2.627	0.769	10.399	2.950	0.025
$La(H_2)_2^{3+}$	2.617	2.636	0.767	10.613	2.886	0.029
$La(H_2)_3^{3+}$	2.647	2.658	0.765	10.710	2.809	0.032
$La(H_2)_4^{3+}$	2.627	2.654	0.762	10.894	2.716	0.036
$La(H_2)_5^{3+}$	2.628	2.678	0.760	10.952	2.631	0.037
$La(H_2)_6^{3+}$	2.650	2.680	0.759	11.085	2.515	0.040
$La(H_2)_7^{3+}$	2.646	2.688	0.757	11.293	2.396	0.043
$La(H_2)_8^{3+}$	2.663	2.697	0.756	11.274	2.277	0.045
$La(H_2)_9^{3+}$	2.674	2.699	0.755	11.787	2.134	0.048
$La(H_2)_{10}^{3+}$	2.678	2.726	0.754	11.255	2.012	0.049
$La(H_2)_{11}^{3+}$	2.681	2.760	0.752	11.166	1.894	0.050
$La(H_2)_{12}^{3+}$	2.730	2.743	0.751	11.860	1.744	0.052
$La(H_2)_{13}^{3+}$	2.663 4.354[a]	2.754	0.750 0.742[d]	8.406	1.742	0.049
$La(H_2)_{14}^{3+}$	2.664 4.309[a] 4.444[b]	2.755	0.751 0.742[d]	8.485	1.741	0.045
$La(H_2)_{15}^{3+}$	2.668 4.345[a] 4.363[b] 4.477[c]	2.760	0.750 0.742[d]	8.478	1.741	0.042

[a,b,c] Represent minimum M-H_2 distance of first, second and third detached H_2 molecule, [d]H-H bond distance in detached H_2 molecules

Table S4. Optimized Bond Lengths ($R_{(M-H)}$, $R_{min(H-H)}$, in Å), HOMO-LUMO Energy Gap (ΔE_{gap}, in eV), Binding Energy as well as Binding Energy per H_2 molecule (BE, in eV) Charge on M (q_M) and Average Charge on H ($q_{avg(H)}$) Atoms (in, e) in $Th(H_2)_{12}^{3+}$ and $M(H_2)_{12}^{4+}$ (M = Th, Pa and U) Calculated with PBE-D3 and B3LYP-D3 Functionals using def-TZVPP Basis Set

System	$R_{(M-H)}$		$R_{min(H-H)}$	ΔE_{gap}	q_M	$q_{avg(H)}$	*BE*	*BE/H₂*
	R_{min}	R_{max}						
$Th(H_2)_{12}^{3+}$								
PBE-D3	2.598	2.767	0.770	0.560	0.734	0.094	-8.710	-0.726
B3LYP-D3	2.750	2.790	0.759	2.904	1.173	0.076	-7.860	-0.655
$Th(H_2)_{12}^{4+}$								
PBE-D3	2.6448	2.656	0.785	4.619	0.828	0.132	-17.471	-1.456
B3LYP-D3	2.6651	2.674	0.773	6.874	1.196	0.117	-16.002	-1.333
$Pa(H_2)_{12}^{4+}$								
PBE-D3	2.591	2.615	0.788	0.415	0.227	0.157	-18.706	-1.559
B3LYP-D3	2.616	2.643	0.775	3.427	0.699	0.138	-17.028	-1.419
$U(H_2)_{12}^{4+}$								
PBE-D3	2.559	2.593	0.790	0.376	0.130	0.161	-19.839	-1.653
B3LYP-D3	2.586	2.613	0.776	3.726	0.621	0.141	-17.962	-1.497

Table S5. Optimized Bond Lengths ($R_{(Ac-H)}$, $R_{min(H-H)}$, in Å), HOMO-LUMO Energy Gap (ΔE_{gap}, in eV), Charge on Ac (q_{Ac}) and Average Charge on H ($q_{avg(H)}$) Atoms (in, e) in $Ac(H_2)_n^{3+}$ (n = 1 - 15) Calculated using PBE-D3/def-TZVPP Method

System	$R_{(Ac-H)}$		$R_{min(H-H)}$	ΔE_{gap}	q_{Ac}	$q_{avg(H)}$
	R_{min}	R_{max}				
$Ac(H_2)^{3+}$	2.716	2.716	0.786	6.694	2.943	0.029
$Ac(H_2)_2^{3+}$	2.725	2.730	0.783	6.777	2.871	0.032
$Ac(H_2)_3^{3+}$	2.727	2.743	0.781	6.868	2.783	0.036
$Ac(H_2)_4^{3+}$	2.725	2.757	0.778	6.924	2.695	0.038
$Ac(H_2)_5^{3+}$	2.730	2.780	0.776	7.017	2.610	0.039
$Ac(H_2)_6^{3+}$	2.756	2.779	0.775	7.178	2.497	0.042
$Ac(H_2)_7^{3+}$	2.756	2.789	0.773	7.286	2.386	0.044
$Ac(H_2)_8^{3+}$	2.772	2.802	0.772	7.263	2.269	0.046
$Ac(H_2)_9^{3+}$	2.784	2.803	0.771	7.615	2.130	0.048
$Ac(H_2)_{10}^{3+}$	2.785	2.819	0.769	7.281	1.985	0.051
$Ac(H_2)_{11}^{3+}$	2.782	2.841	0.768	7.316	1.820	0.054
$Ac(H_2)_{12}^{3+}$	2.807	2.818	0.767	8.012	1.635	0.057
$Ac(H_2)_{13}^{3+}$	2.741 4.415[a]	2.831	0.767 0.758[d]	4.665	1.638	0.052
$Ac(H_2)_{14}^{3+}$	2.741 4.370[a] 4.378[b]	2.830	0.767 0.758[d]	4.744	1.630	0.049
$Ac(H_2)_{15}^{3+}$	2.742 4.413[a] 4.425[c] 4.430[c]	2.845	0.767 0.757[d]	4.721	1.627	0.046

[a,b,c] Represent minimum M-H_2 distance of first, second and third detached H_2 molecule, [d]H-H bond distance in detached H_2 molecule

Table S6. Optimized Bond Lengths ($R_{(La-H)}$, $R_{min(H-H)}$, in Å), HOMO-LUMO Energy Gap (ΔE_{gap}, in eV), Charge on La (q_{La}) and Average Charge on H ($q_{avg(H)}$) Atoms (in, e) in $La(H_2)_n^{3+}$ (n = 1 - 15) Calculated using PBE-D3/Def-TZVPP Method

System	$R_{(La-H)}$		$R_{min(H-H)}$	ΔE_{gap}	q_{La}	$q_{avg(H)}$
	R_{min}	R_{max}				
$La(H_2)^{3+}$	2.625	2.625	0.791	5.463	2.919	0.041
$La(H_2)_2^{3+}$	2.612	2.634	0.787	5.687	2.823	0.044
$La(H_2)_3^{3+}$	2.616	2.665	0.784	5.813	2.717	0.047
$La(H_2)_4^{3+}$	2.620	2.661	0.781	5.967	2.593	0.051
$La(H_2)_5^{3+}$	2.614	2.693	0.778	6.121	2.478	0.052
$La(H_2)_6^{3+}$	2.641	2.681	0.777	6.323	2.328	0.056
$La(H_2)_7^{3+}$	2.640	2.685	0.775	6.465	2.186	0.058
$La(H_2)_8^{3+}$	2.653	2.706	0.774	6.429	2.048	0.060
$La(H_2)_9^{3+}$	2.674	2.705	0.773	6.867	1.876	0.062
$La(H_2)_{10}^{3+}$	2.676	2.728	0.771	6.398	1.726	0.064
$La(H_2)_{11}^{3+}$	2.666	2.768	0.770	6.363	1.574	0.065
$La(H_2)_{12}^{3+}$	2.728	2.741	0.769	6.958	1.392	0.067
$La(H_2)_{13}^{3+}$	2.630 4.374[a]	2.770	0.768 0.759[d]	3.691	1.391	0.062
$La(H_2)_{14}^{3+}$	2.636 4.335[a] 4.342[b]	2.758	0.768 0.758[d]	3.741	1.389	0.0575
$La(H_2)_{15}^{3+}$	2.627 4.379[a] 4.373[b] 4.385[c]	2.788	0.768 0.758[d]	3.744	1.392	0.0535

[a,b,c] Represent minimum M-H_2 distance of first, second and third detached H_2 molecule, [d]H-H bond distance in detached H_2 molecule

Table S7. Optimized Bond Lengths ($R_{(Ac-H)}$, $R_{min(H-H)}$, in Å), HOMO-LUMO Energy Gap (ΔE_{gap}, in eV), Charge on Ac (q_{Ac}) and Average Charge on H ($q_{avg(H)}$) Atoms (in, e) in $Ac(H_2)_n^{3+}$ (n = 1 - 15) Calculated using B3LYP-D3/def-TZVPP Method

System	$R_{(Ac-H)}$		$R_{min(H-H)}$	ΔE_{gap}	q_{Ac}	$q_{avg(H)}$
	R_{min}	R_{max}				
$Ac(H_2)^{3+}$	2.734	...	0.774	8.575	2.954	0.023
$Ac(H_2)_2^{3+}$	2.761	2.764	0.772	8.580	2.897	0.026
$Ac(H_2)_3^{3+}$	2.746	2.760	0.770	8.758	2.822	0.030
$Ac(H_2)_4^{3+}$	2.749	2.773	0.768	8.825	2.749	0.031
$Ac(H_2)_5^{3+}$	2.755	2.796	0.765	8.825	2.680	0.032
$Ac(H_2)_6^{3+}$	2.779	2.797	0.764	9.015	2.581	0.035
$Ac(H_2)_7^{3+}$	2.778	2.808	0.763	9.179	2.483	0.037
$Ac(H_2)_8^{3+}$	2.793	2.821	0.761	9.142	2.380	0.039
$Ac(H_2)_9^{3+}$	2.803	2.822	0.761	9.556	2.257	0.041
$Ac(H_2)_{10}^{3+}$	2.810	2.834	0.759	9.213	2.132	0.043
$Ac(H_2)_{11}^{3+}$	2.802	2.862	0.758	9.219	1.996	0.046
$Ac(H_2)_{12}^{3+}$	2.832	2.846	0.757	9.918	1.844	0.048
$Ac(H_2)_{13}^{3+}$	2.774 4.473[a]	2.852	0.757 0.749[d]	6.433	1.841	0.045
$Ac(H_2)_{14}^{3+}$	2.771 4.409[a] 4.419[b]	2.854	0.757 0.749[d]	6.535	1.834	0.042
$Ac(H_2)_{15}^{3+}$	2.775 4.452[a] 4.470[b] 4.472[c]	2.858	0.757 0.749[d]	6.502	1.830	0.039

[a,b,c] Represent minimum M-H_2 distance of first, second and third detached H_2 molecule, [d]H-H bond distance in detached H_2 molecule

Table S8. Optimized Bond Lengths ($R_{(La-H)}$, $R_{min(H-H)}$, in Å), HOMO-LUMO Energy Gap (ΔE_{gap}, in eV), Charge on La (q_{La}) and Average Charge on H ($q_{avg(H)}$) Atoms (in, e) in $La(H_2)_n^{3+}$ (n = 1 - 15) Calculated using B3LYP-D3/def-TZVPP Method

System	$R_{(La-H)}$		$R_{min(H-H)}$	ΔE_{gap}	q_{La}	$q_{avg(H)}$
	R_{min}	R_{max}				
$La(H_2)^{3+}$	2.645		0.778	7.313	2.936	0.032
$La(H_2)_2^{3+}$	2.634	2.651	0.775	7.549	2.858	0.035
$La(H_2)_3^{3+}$	2.659	2.682	0.773	7.658	2.768	0.039
$La(H_2)_4^{3+}$	2.637	2.676	0.770	7.854	2.664	0.042
$La(H_2)_5^{3+}$	2.640	2.704	0.768	7.934	2.570	0.043
$La(H_2)_6^{3+}$	2.669	2.702	0.766	8.077	2.443	0.046
$La(H_2)_7^{3+}$	2.663	2.707	0.765	8.295	2.315	0.049
$La(H_2)_8^{3+}$	2.681	2.719	0.763	8.279	2.192	0.051
$La(H_2)_9^{3+}$	2.692	2.720	0.762	8.742	2.040	0.053
$La(H_2)_{10}^{3+}$	2.699	2.751	0.761	8.241	1.913	0.054
$La(H_2)_{11}^{3+}$	2.699	2.787	0.759	8.166	1.792	0.055
$La(H_2)_{12}^{3+}$	2.748	2.771	0.758	8.759	1.638	0.057
$La(H_2)_{13}^{3+}$	2.669, 4.440[a]	2.792	0.758 0.749[d]	5.308	1.638	0.052
$La(H_2)_{14}^{3+}$	2.669 4.378 [a] 4.386[b]	2.786	0.757 0.749[d]	5.401	1.635	0.049
$La(H_2)_{15}^{3+}$	2.675, 4.427[a] 4.438[b] 4.439[c]	2.802	0.757 0.749[d]	5.378	1.636	0.045

[a,b,c] Represent minimum M-H_2 distance of first, second and third detached H_2 molecule, [d]H-H distance of detached H_2 molecule

Table S9. Calculated Value of Relative Energy (RE, in eV) of Mixed Species ($[Ac(H)_2(H_2)_y^{3+}]$ and $[Ac(H)_4(H_2)_y^{3+}]$, where y = 1, 2, 9-10) with Respect to the Corresponding Similar Composition $Ac(H_2)_n^{3+}$ System and their Binding Energy (BE, in eV) at BHLYP/def-TZVPP Level of Theory. Binding Energy of $Ac(H_2)_n^{3+}$ (BE, in eV) Systems is Provided within Parenthesis

Systems	RE	BE
$Ac(H)_2(H_2)^{3+}$	6.454	0.190 (-1.563)
$Ac(H)_2(H_2)_2^{3+}$	6.529	-0.443 (-2.270)
$Ac(H)_4(H_2)^{3+}$	13.153	1.472 (-2.270)
$Ac(H)_4(H_2)_2^{3+}$	13.271	0.927 (-2.924)
$Ac(H)_2(H_2)_9^{3+}$	6.854	-3.919 (-6.064)
$Ac(H)_2(H_2)_{10}^{3+}$	6.813	-4.324 (-6.436)

Table S10. Calculated Value of Optimized Bond Lengths (in Å), HOMO-LUMO Gap (ΔE_{gap}, in eV), Charge on Metal ion (q_{Ac}, in e) and Average NPA Charge on H atom (q_H, in e) and on H atom of H_2 Molecule (q_{H2}, in e) in Mixed Species at BHLYP-D3/def-TZVPP Level of Theory

Systems	$R_{(M-H)}$	$R_{min(M-H2)}$	$R_{max(M-H2)}$	$R_{(H-H)}$	ΔE_{gap}	q_{Ac}	$q_H(q_{H2})$
$Ac(H)_2(H_2)^{3+}$	2.412-2.421	2.750	2.750	0.761	3.732	2.423	0.26 (0.03)
$Ac(H)_2(H_2)_2^{3+}$	2.382-2.384	2.753	2.764	0.759	3.828	2.304	0.28 (0.03)
$Ac(H)_4(H_2)^{3+}$	2.322-2.323	2.772	2.773	0.757	3.951	1.522	0.35 (0.04)
$Ac(H)_4(H_2)_2^{3+}$	2.285-2.302	2.774	2.800	0.756	4.092	1.382	0.36 (0.04)
$Ac(H)_2(H_2)_9^{3+}$	2.182-2.183	2.789	2.845	0.752	4.445	1.548	0.29 (0.05)
$Ac(H)_2(H_2)_{10}^{3+}$	2.166-2.167	2.819	2.825	0.751	4.538	1.402	0.29 (0.05)

Table S11. Binding Energy and Binding Energy per H_2 Molecule (BE, in eV) of $Ac(H_2)_n^{3+}$ (n = 1 - 12) Calculated using PBE-D3, B3LYP-D3, BHLYP-D3 Functionals with def-TZVPP Basis Set.

Systems	PBE-D3		B3LYP-D3		BHLYP-D3	
	BE	BE/H_2	BE	BE/H_2	BE	BE/H_2
$Ac(H_2)^{3+}$	-0.939	-0.939	-0.845	-0.845	-0.809	-0.809
$Ac(H_2)_2^{3+}$	-1.805	-0.902	-1.628	-0.814	-1.563	-0.781
$Ac(H_2)_3^{3+}$	-2.610	-0.870	-2.354	-0.785	-2.270	-0.757
$Ac(H_2)_4^{3+}$	-3.343	-0.836	-3.020	-0.755	-2.925	-0.731
$Ac(H_2)_5^{3+}$	-4.001	-0.800	-3.614	-0.723	-3.514	-0.703
$Ac(H_2)_6^{3+}$	-4.663	-0.777	-4.217	-0.703	-4.109	-0.685
$Ac(H_2)_7^{3+}$	-5.248	-0.750	-4.743	-0.678	-4.637	-0.662
$Ac(H_2)_8^{3+}$	-5.811	-0.726	-5.256	-0.657	-5.154	-0.644
$Ac(H_2)_9^{3+}$	-6.350	-0.706	-5.744	-0.638	-5.647	-0.627
$Ac(H_2)_{10}^{3+}$	-6.809	-0.681	-6.150	-0.615	-6.064	-0.606
$Ac(H_2)_{11}^{3+}$	-7.223	-0.657	-6.504	-0.591	-6.436	-0.585
$Ac(H_2)_{12}^{3+}$	-7.663	-0.639	-6.895	-0.575	-6.838	-0.570

Table S12. Binding Energy and Binding Energy per H_2 Molecule (BE, in eV) of $La(H_2)_n^{3+}$ (n = 1 - 12) Calculated using PBE-D3, B3LYP-D3, BHLYP-D3 Functionals with def-TZVPP Basis Set.

LaH2_n	PBE-D3		B3LYP-D3		BHLYP-D3	
	BE	BE/H_2	BE	BE/H_2	BE	BE/H_2
$La(H_2)^{3+}$	-1.053	-1.053	-0.951	-0.951	-0.898	-0.898
$La(H_2)_2^{3+}$	-2.010	-1.005	-1.817	-0.909	-1.727	-0.863
$La(H_2)_3^{3+}$	-2.881	-0.960	-2.621	-0.874	-2.498	-0.833
$La(H_2)_4^{3+}$	-3.694	-0.924	-3.352	-0.838	-3.214	-0.804
$La(H_2)_5^{3+}$	-4.420	-0.884	-4.010	-0.802	-3.863	-0.773
$La(H_2)_6^{3+}$	-5.133	-0.855	-4.671	-0.779	-4.513	-0.752
$La(H_2)_7^{3+}$	-5.775	-0.825	-5.255	-0.751	-5.093	-0.728
$La(H_2)_8^{3+}$	-6.389	-0.799	-5.827	-0.728	-5.667	-0.708
$La(H_2)_9^{3+}$	-6.979	-0.775	-6.368	-0.708	-6.211	-0.690
$La(H_2)_{10}^{3+}$	-7.431	-0.743	-6.779	-0.678	-6.631	-0.663
$La(H_2)_{11}^{3+}$	-7.816	-0.711	-7.118	-0.647	-6.985	-0.635
$La(H_2)_{12}^{3+}$	-8.260	-0.688	-7.517	-0.626	-7.399	-0.617

Table S13. Basis Set Superimposition Error (BSSE, in eV) Calculated Using BHLYP-D3/def-TZVPP Method.

System	BSSE	System	BSSE
$Ac(H_2)^{3+}$	0.001	$U(H_2)_{12}^{4+}$	0.023
$Ac(H_2)_2^{3+}$	0.002	$Th(H_2)_{12}^{3+}$	0.019
$Ac(H_2)_3^{3+}$	0.003	$Th(H_2)_{12}^{4+}$	0.025
$Ac(H_2)_4^{3+}$	0.004	$La(H_2)_{12}^{3+}$	0.012
$Ac(H_2)_5^{3+}$	0.006		
$Ac(H_2)_6^{3+}$	0.006		
$Ac(H_2)_7^{3+}$	0.008		
$Ac(H_2)_8^{3+}$	0.009		
$Ac(H_2)_9^{3+}$	0.010		
$Ac(H_2)_{10}^{3+}$	0.012		
$Ac(H_2)_{11}^{3+}$	0.014		
$Ac(H_2)_{12}^{3+}$	0.015		

Table S14. Binding Energy (BE, in eV) and Binding Energy per H_2 Molecule (BE/H_2, in eV) Calculated Using MP2 and NEO-MP2 Method using def2-TZVPP Basis set for H and CRENBL Basis set of Ac. (DZSPDN Nuclear Basis Set is used for the Quantum Hydrogen)

System	MP2		NEO-MP2		BE_Error	BE_Error/H_2
	BE	BE/H_2	BE	BE/H_2		
$Ac(H_2)^{3+}$	-0.698	-0.698	-0.784	-0.784	0.086	0.086
$Ac(H_2)_2^{3+}$	-1.369	-0.684	-1.531	-0.765	0.162	0.081
$Ac(H_2)_3^{3+}$	-2.002	-0.667	-2.230	-0.743	0.228	0.076
$Ac(H_2)_4^{3+}$	-2.609	-0.652	-2.894	-0.723	0.285	0.071
$Ac(H_2)_5^{3+}$	-3.155	-0.631	-3.483	-0.697	0.328	0.066
$Ac(H_2)_6^{3+}$	-3.723	-0.621	-4.100	-0.683	0.377	0.063
$Ac(H_2)_7^{3+}$	-4.219	-0.603	-4.630	-0.661	0.410	0.059

Table S15. Calculated Values of Bond Critical Point Electron Density (ρ in e a_0^{-3}), Laplacian of Electron Density ($\nabla^2\rho$ in e a_0^{-5}), Local Electron Energy Density (E_d in au), and Ratio of Local Electron Kinetic Energy Density and Electron Density ($G(r)/\rho$ in au) of $Ac(H_2)_n^{3+}$ (n = 1 - 12), $Th(H_2)_{12}^{3+}$, $Th(H_2)_{12}^{4+}$, $Pa(H_2)_{12}^{4+}$, $U(H_2)_{12}^{4+}$ Clusters as obtained by using BHLYP-D3/def-TZVPP Method along with Small Core ECP Employed with EDF

System	Bond	ρ	$\nabla^2\rho$	G(r)[c]	V(r)[d]	$E_d(r)$	G(r)/ρ	Type[e]
$Ac(H_2)^{3+}$	Ac-H	0.0238	0.0471	0.0126	-0.0133	-0.0008	0.529	D
	H-H	0.2499	-1.0860	0.0001	-0.2716	-0.2716	0.000	A
$Ac(H_2)_2^{3+}$	Ac-H	0.0220	0.0454	0.0118	-0.0122	-0.0004	0.536	D
	H-H	0.2514	-1.0960	0.0000	-0.2741	-0.2741	0.000	A
$Ac(H_2)_3^{3+}$	Ac-H	0.0223	0.0474	0.0122	-0.0126	-0.0004	0.547	D
	H-H	0.2531	-1.1070	0.0001	-0.2769	-0.2768	0.000	A
$Ac(H_2)_4^{3+}$	Ac-H	0.0216	0.0472	0.0120	-0.0122	-0.0002	0.556	D
	H-H	0.2547	-1.1180	0.0001	-0.2795	-0.2795	0.000	A
$Ac(H_2)_5^{3+}$	Ac-H	0.0211	0.0470	0.0118	-0.0119	-0.0001	0.560	D
	H-H	0.2565	-1.1300	0.0001	-0.2826	-0.2826	0.000	A
$Ac(H_2)_6^{3+}$	Ac-H	0.0204	0.0468	0.0116	-0.0116	0.0001	0.569	D
	H-H	0.2573	-1.1340	0.0001	-0.2837	-0.2837	0.000	A
$Ac(H_2)_7^{3+}$	Ac-H	0.0197	0.0464	0.0114	-0.0112	0.0002	0.579	D
	H-H	0.2584	-1.1420	0.0001	-0.2856	-0.2856	0.000	A
$Ac(H_2)_8^{3+}$	Ac-H	0.0196	0.0469	0.0115	-0.0113	0.0002	0.587	D
	H-H	0.2595	-1.1490	0.0001	-0.2874	-0.2875	0.000	A
$Ac(H_2)_9^{3+}$	Ac-H	0.0192	0.0469	0.0114	-0.0111	0.0003	0.594	D
	H-H	0.2604	-1.1550	0.0001	-0.2890	-0.2890	0.000	A
$Ac(H_2)_{10}^{3+}$	Ac-H	0.0190	0.0471	0.0114	-0.0110	0.0004	0.599	D
	H-H	0.2617	-1.1640	0.0001	-0.2911	-0.2910	0.000	A
$Ac(H_2)_{11}^{3+}$	Ac-H	0.0181	0.0454	0.0109	-0.0105	0.0004	0.604	D
	H-H	0.2631	-1.1730	0.0002	-0.2935	-0.2934	0.001	A
$Ac(H_2)_{12}^{3+}$	Ac-H	0.0177	0.0452	0.0108	-0.0103	-0.0005	0.611	D
	H-H	0.2639	-1.1780	0.0002	-0.2949	-0.2947	0.001	A
$Th(H_2)_{12}^{3+}$	Th-H	0.0192	0.0489	0.0124	-0.0125	-0.0002	0.643	D
	H-H	0.2627	-1.1680	0.0003	-0.2928	-0.2925	0.001	A
$Th(H_2)_{12}^{4+}$	Th-H	0.0281	0.0481	0.0147	-0.0173	-0.0027	0.523	D
	H-H	0.2510	-1.0910	0.0004	-0.2736	-0.2732	0.002	A
$Pa(H_2)_{12}^{4+}$	Pa-H	0.0291	0.0546	0.0164	-0.0191	-0.0027	0.562	D
	H-H	0.2503	-1.0870	0.0004	-0.2727	-0.2723	0.002	A
$U(H_2)_{12}^{4+}$	Pa-H	0.0309	0.0572	0.0174	-0.0205	-0.0031	0.563	D
	H-H	0.2494	-1.0820	0.0005	-0.2716	-0.2711	0.002	A

Table S16. Calculated Values of Bond Critical Point Electron Density (ρ in e a_0^{-3}), Laplacian of Electron Density ($\nabla^2\rho$ in e a_0^{-5}), Local Electron Energy Density (E_d in au), and Ratio of Local Electron Kinetic Energy Density and Electron Density ($G(r)/\rho$ in au) of $La(H_2)_n^{3+}$ (n = 1 - 12) Clusters as obtained by using BHLYP-D3/def-TZVPP Method along with Small Core ECP Employed with EDF

System	Bond	ρ	$\nabla^2\rho$	G(r)[c]	V(r)[d]	$E_d(r)$	G(r)/ρ	Type[e]
$La(H_2)^{3+}$	La-H	0.0249	0.0598	0.0145	-0.0141	0.0004	0.582	D
	H-H	0.2483	-2.1450	0.0000	-0.2682	-0.2682	0.000	A
$La(H_2)_2^{3+}$	La-H	0.0245	0.0609	0.0147	-0.0141	0.0005	0.598	D
	H-H	0.2499	-2.0820	0.0000	-0.2707	-0.2706	0.000	A
$La(H_2)_3^{3+}$	La-H	0.0230	0.0588	0.0139	-0.0131	0.0007	0.605	D
	H-H	0.2517	-1.0940	0.0000	-0.2737	-0.2736	0.000	A
$La(H_2)_4^{3+}$	La-H	0.0232	0.0612	0.0145	-0.0136	0.0008	0.623	D
	H-H	0.2537	-1.1070	0.0001	-0.2769	-0.2768	0.000	A
$La(H_2)_5^{3+}$	La-H	0.0223	0.0608	0.0142	-0.0132	0.0009	0.636	D
	H-H	0.2556	-1.1200	0.0001	-0.2803	-0.2801	0.000	A
$La(H_2)_6^{3+}$	La-H	0.0217	0.0600	0.0139	-0.0129	0.0010	0.642	D
	H-H	0.2565	-1.1260	0.0001	-0.2817	-0.2816	0.000	A
$La(H_2)_7^{3+}$	La-H	0.0207	0.0596	0.0136	-0.0124	0.0012	0.657	D
	H-H	0.2580	-1.1350	0.0001	-0.2842	-0.2840	0.000	A
$La(H_2)_8^{3+}$	La-H	0.0205	0.0595	0.0136	-0.0123	0.0012	0.663	D
	H-H	0.2592	-1.1450	0.0001	-0.2865	-0.2864	0.000	A
$La(H_2)_9^{3+}$	La-H	0.0199	0.0585	0.0133	-0.0120	0.0013	0.668	D
	H-H	0.2599	-1.1490	0.0001	-0.2876	-0.2875	0.000	A
$La(H_2)_{10}^{3+}$	La-H	0.0190	0.0573	0.0129	-0.0115	0.0014	0.679	D
	H-H	0.2614	-1.1600	0.0001	-0.2902	-0.2901	0.001	A
$La(H_2)_{11}^{3+}$	La-H	0.0185	0.0566	0.0127	-0.0113	0.0014	0.687	D
	H-H	0.2627	-1.1680	0.0002	-0.2925	-0.2923	0.001	A
$La(H_2)_{12}^{3+}$	La-H	0.0177	0.0551	0.0123	-0.0108	0.0015	0.694	D
	H-H	0.2638	-1.1750	0.0002	-0.2944	-0.2942	0.001	A

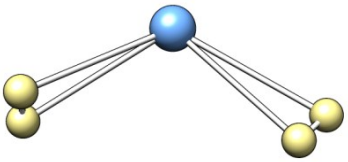
Table S17. Calculated Value of Optimized Bond Lengths (in Å), Binding Energy (BE, in eV), Binding Energy Per Hydrogen Molecule (BE/H_2, in eV), Energy Gain on H_2 addition (EG, in eV), HOMO-LUMO Energy Gap ($\Delta E_{gap,}$ in eV) and Voronoi Deformation Density Charge (VDD) on Metal ion (q_M, in e) in $Ac(H_2)_n^{3+}$ (n = 1-12) and $M(H_2)_{12}^{3+/4+}$ (M = La, Th, U) System using Scalar Relativistic ZORA Approach at PBE-D3BJ/TZ2P Level of Theory with 4f-Frozen Core

System	$R_{min(M-H)}$	$R_{max(M-H)}$	$R_{(H-H)}$	BE	BE/H_2	EG	ΔE_{gap}	q_M
$Ac(H_2)^{3+}$	2.713	2.713	0.785	-0.961	-0.961	-0.961	6.673	2.352
$Ac(H_2)_2^{3+}$	2.769	2.776	0.782	-1.845	-0.922	-0.883	6.527	1.815
$Ac(H_2)_3^{3+}$	2.736	2.743	0.780	-2.633	-0.878	-0.788	6.840	1.305
$Ac(H_2)_4^{3+}$	2.750	2.751	0.778	-3.395	-0.849	-0.762	6.900	0.839
$Ac(H_2)_5^{3+}$	2.738	2.786	0.775	-4.055	-0.811	-0.661	6.912	1.004
$Ac(H_2)_6^{3+}$	2.760	2.782	0.774	-4.735	-0.789	-0.680	7.132	0.606
$Ac(H_2)_7^{3+}$	2.762	2.791	0.773	-5.342	-0.763	-0.607	7.257	0.525
$Ac(H_2)_8^{3+}$	2.778	2.811	0.772	-5.926	-0.741	-0.584	7.192	0.454
$Ac(H_2)_9^{3+}$	2.789	2.810	0.771	-6.488	-0.721	-0.562	7.516	0.379
$Ac(H_2)_{10}^{3+}$	2.794	2.825	0.769	-6.973	-0.697	-0.485	7.191	0.356
$Ac(H_2)_{11}^{3+}$	2.790	2.841	0.768	-7.420	-0.675	-0.447	7.238	0.350
$Ac(H_2)_{12}^{3+}$	2.823	2.828	0.768	-7.883	-0.657	-0.463	7.915	0.320
$La(H_2)_{12}^{3+}$	2.714	2.721	0.769	-9.041	-0.753	...	5.075	0.324
$Th(H_2)_{12}^{3+}$	2.482	2.813	0.769	-9.146	-0.762	...	0.556	0.119
$Th(H_2)_{12}^{4+}$	2.645	2.654	0.785	-17.498	-1.458	...	4.847	0.382
$U(H_2)_{12}^{4+}$	2.550	2.584	0.789	-20.375	-1.698	...	0.335	0.528

Table S18. Calculated Value of Optimized Bond Lengths (in Å), Binding Energy (BE, in eV), Binding Energy Per Hydrogen Molecule (BE/H_2, in eV), Energy Gain on H_2 addition (EG, in eV), HOMO-LUMO Energy Gap ($\Delta E_{gap,}$ in eV) and Voronoi Deformation Density Charge (VDD) on Metal ion (q_M, in e) in $Ac(H_2)_n^{3+}$ (n = 1-12) and $M(H_2)_{12}^{3+/4+}$ (M = La, Th, U) System using Scalar Relativistic ZORA Approach with All Electron Basis Set at PBE-D3BJ/TZ2P Level of Theory

System	$R_{min(M-H)}$	$R_{max(M-H)}$	$R_{(H-H)}$	BE	BE/H_2	EG	ΔE_{gap}	q_M
$Ac(H_2)^{3+}$	2.714	2.714	0.785	-0.971	-0.971	-0.971	6.719	2.355
$Ac(H_2)_2^{3+}$	2.768	2.777	0.783	-1.864	-0.932	-0.893	6.598	1.838
$Ac(H_2)_3^{3+}$	2.732	2.739	0.780	-2.663	-0.888	-0.799	6.912	1.300
$Ac(H_2)_4^{3+}$	2.745	2.747	0.778	-3.434	-0.859	-0.772	6.964	0.840
$Ac(H_2)_5^{3+}$	2.734	2.786	0.776	-4.100	-0.820	-0.665	7.004	1.009
$Ac(H_2)_6^{3+}$	2.757	2.776	0.774	-4.792	-0.799	-0.692	7.134	0.596
$Ac(H_2)_7^{3+}$	2.760	2.787	0.773	-5.405	-0.772	-0.614	7.312	0.524
$Ac(H_2)_8^{3+}$	2.772	2.809	0.771	-5.993	-0.749	-0.588	7.245	0.456
$Ac(H_2)_9^{3+}$	2.780	2.808	0.770	-6.559	-0.729	-0.566	7.502	0.382
$Ac(H_2)_{10}^{3+}$	2.784	2.820	0.769	-7.056	-0.706	-0.497	7.245	0.356
$Ac(H_2)_{11}^{3+}$	2.783	2.831	0.768	-7.508	-0.683	-0.452	7.296	0.353
$Ac(H_2)_{12}^{3+}$	2.815	2.820	0.767	-7.976	-0.665	-0.468	7.984	0.323
$La(H_2)_{12}^{3+}$	2.710	2.717	0.770	-9.076	-0.756	...	5.077	0.331
$Th(H_2)_{12}^{3+}$	2.531	2.791	0.770	-9.187	-0.766	...	0.569	0.098
$Th(H_2)_{12}^{4+}$	2.640	2.649	0.785	-17.612	-1.468	...	4.708	0.380
$U(H_2)_{12}^{4+}$	2.551	2.578	0.789	-20.386	-1.699	...	0.342	0.531

1



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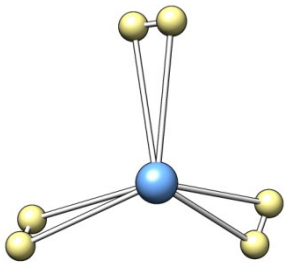

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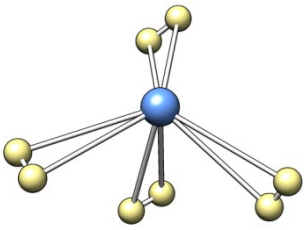

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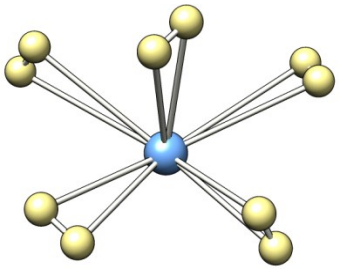

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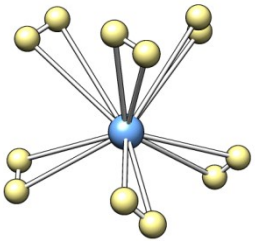

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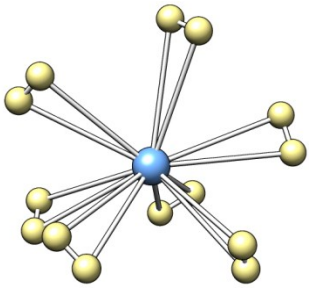

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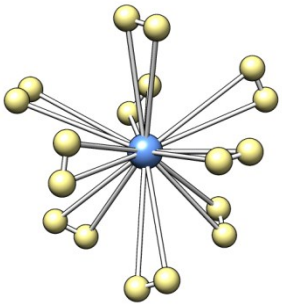

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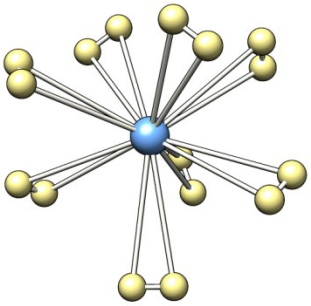

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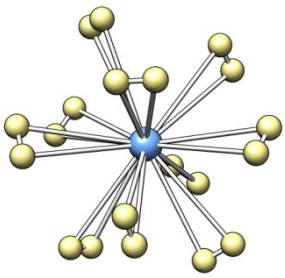

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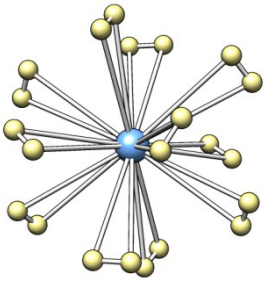

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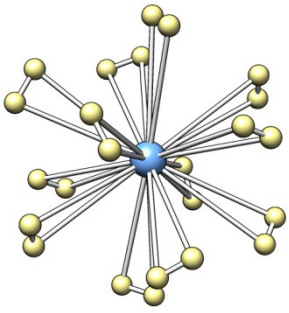

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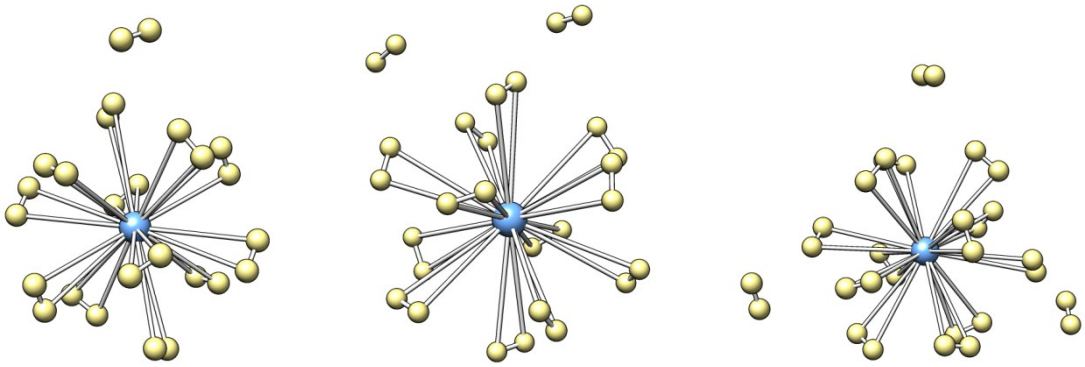


Figure S1. . Optimized structures of $M(H_2)_n^{3+}$ (n = 1-15) using BHLYP-D3/def-TZVPP method

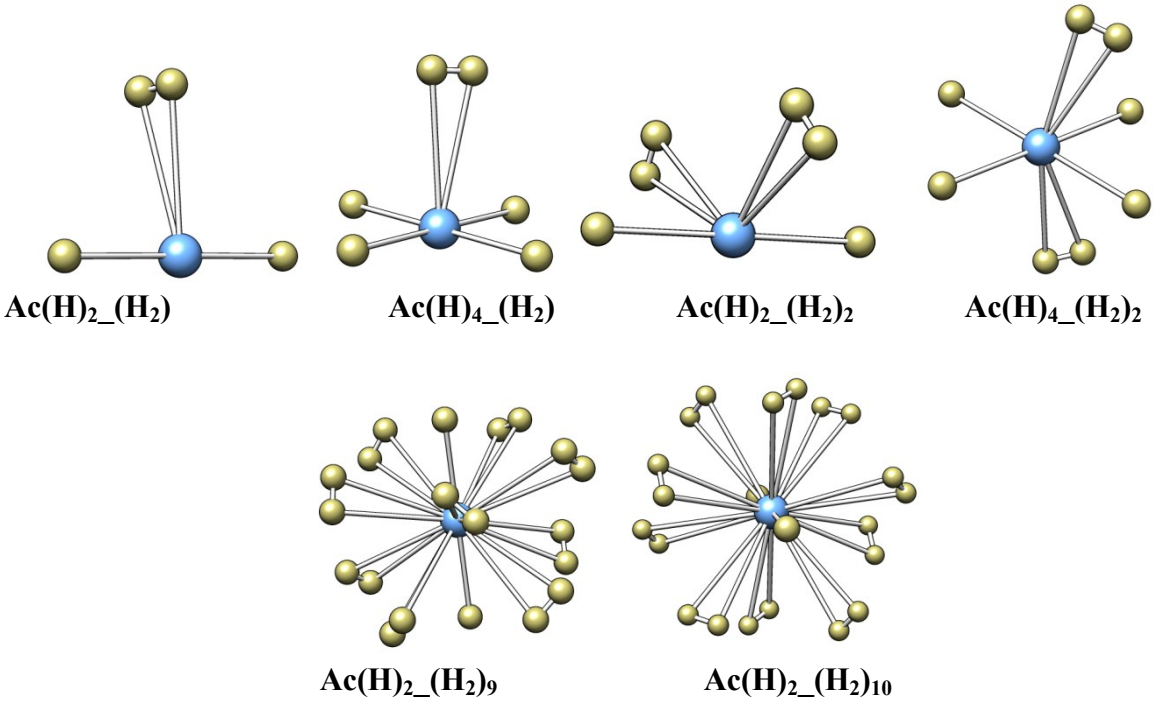


Figure S2. . Optimized structures of $Ac(H)_2(H_2)_y^{3+}$ and $Ac(H)_4(H_2)_y^{3+}$ systems (where y = 1, 2, 9-10) using BHLYP-D3/def-TZVPP method

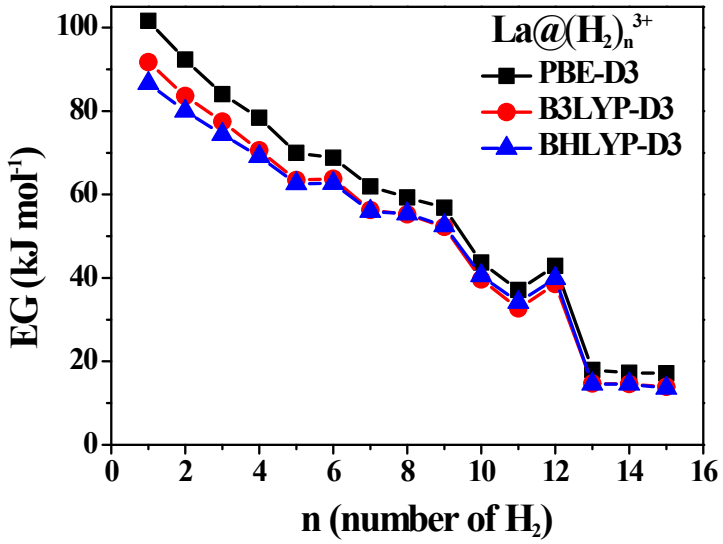


Figure S3. Energy Gain (EG, kJ mol^{-1}) of La$(H_2)_n^{3+}$ (n = 1 - 15) systems on addition of hydrogen molecule in La$(H_2)_{n-1}^{3+}$ systems .

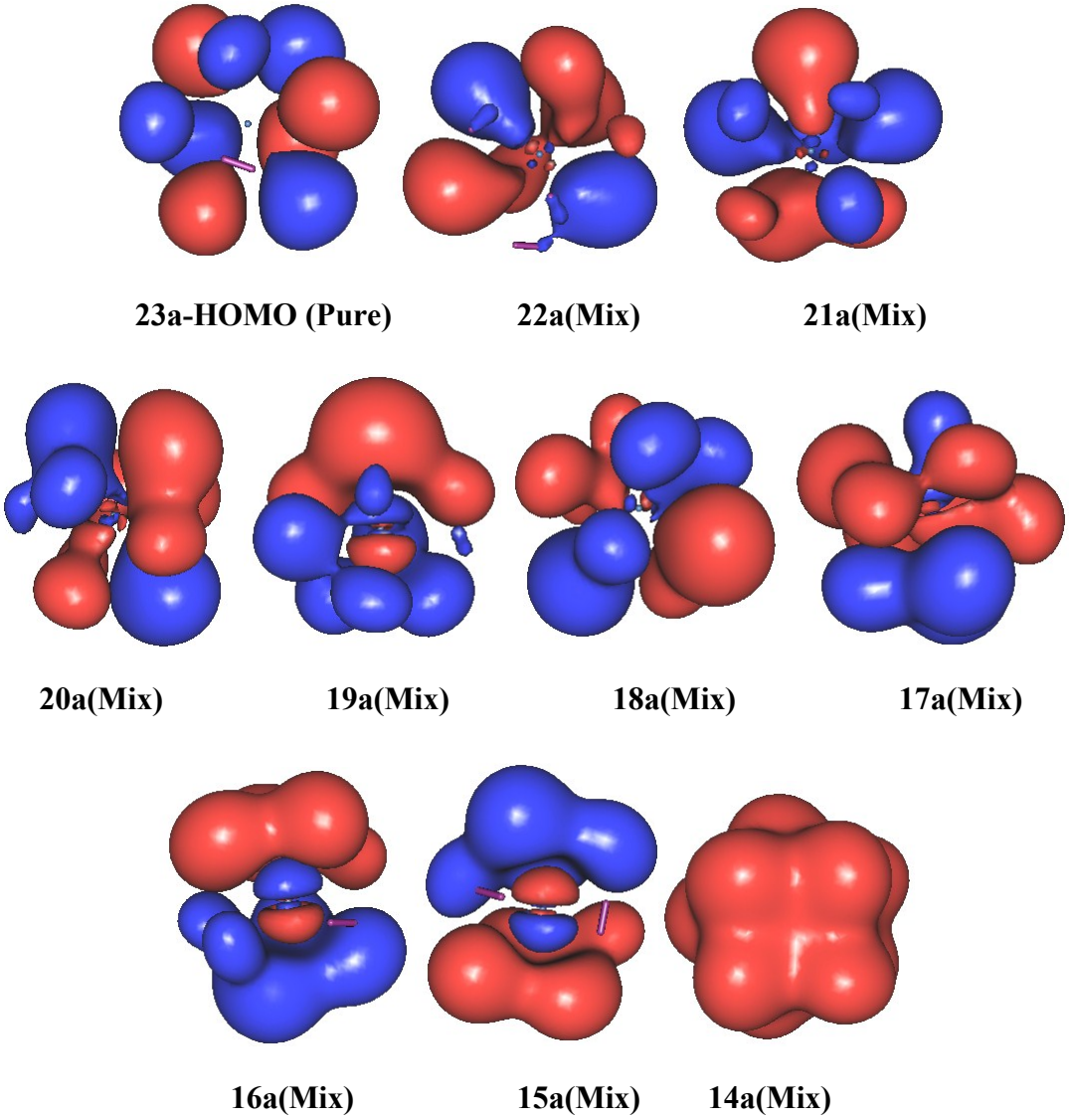


Figure S4. MOs Picture of $Ac(H_2)_{10}^{3+}$ using BHLYP-D3/def-TZVPP method

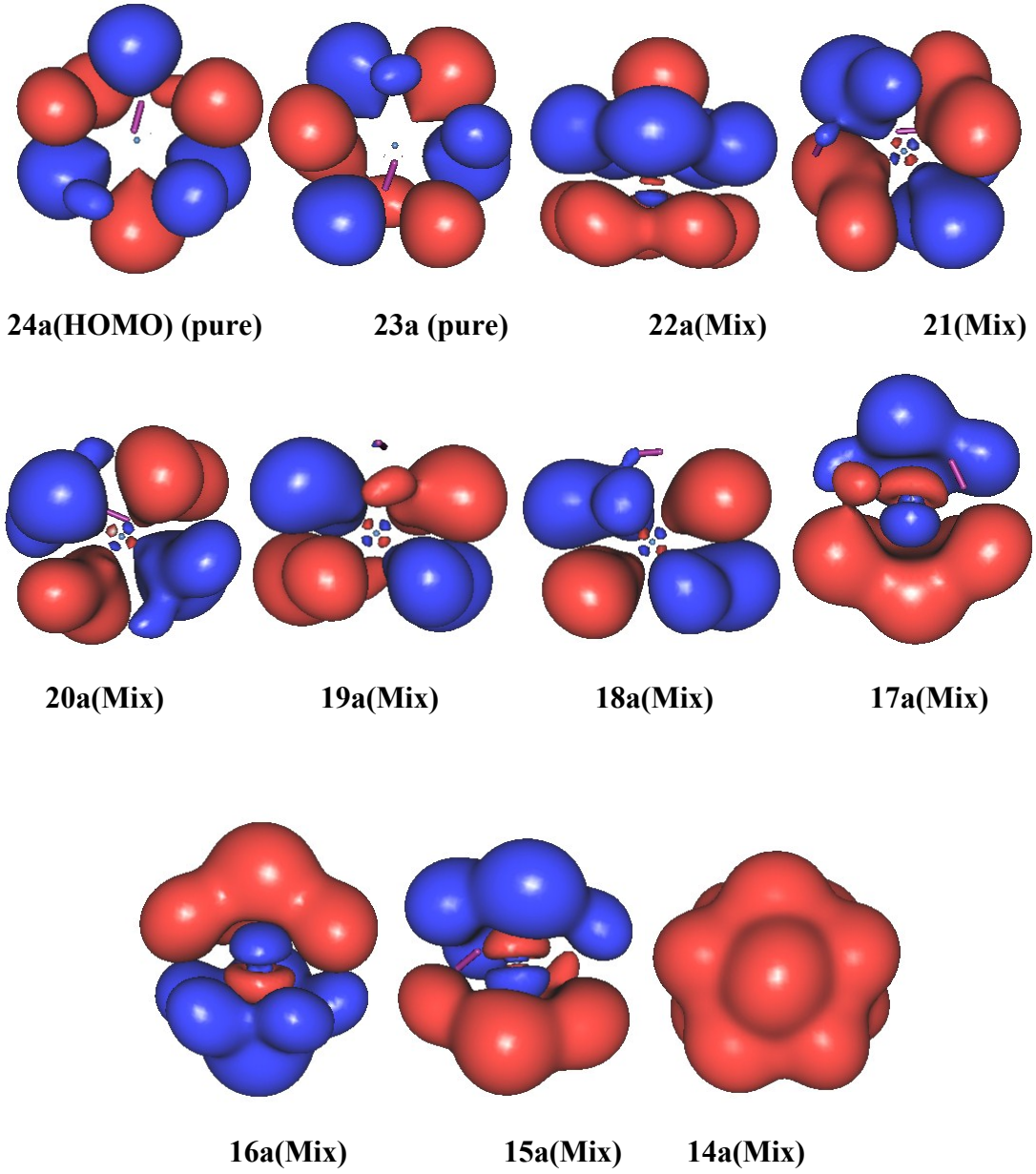


Figure S5. MOs Picture of $Ac(H_2)_{11}^{3+}$ using BHLYP-D3/def-TZVPP method

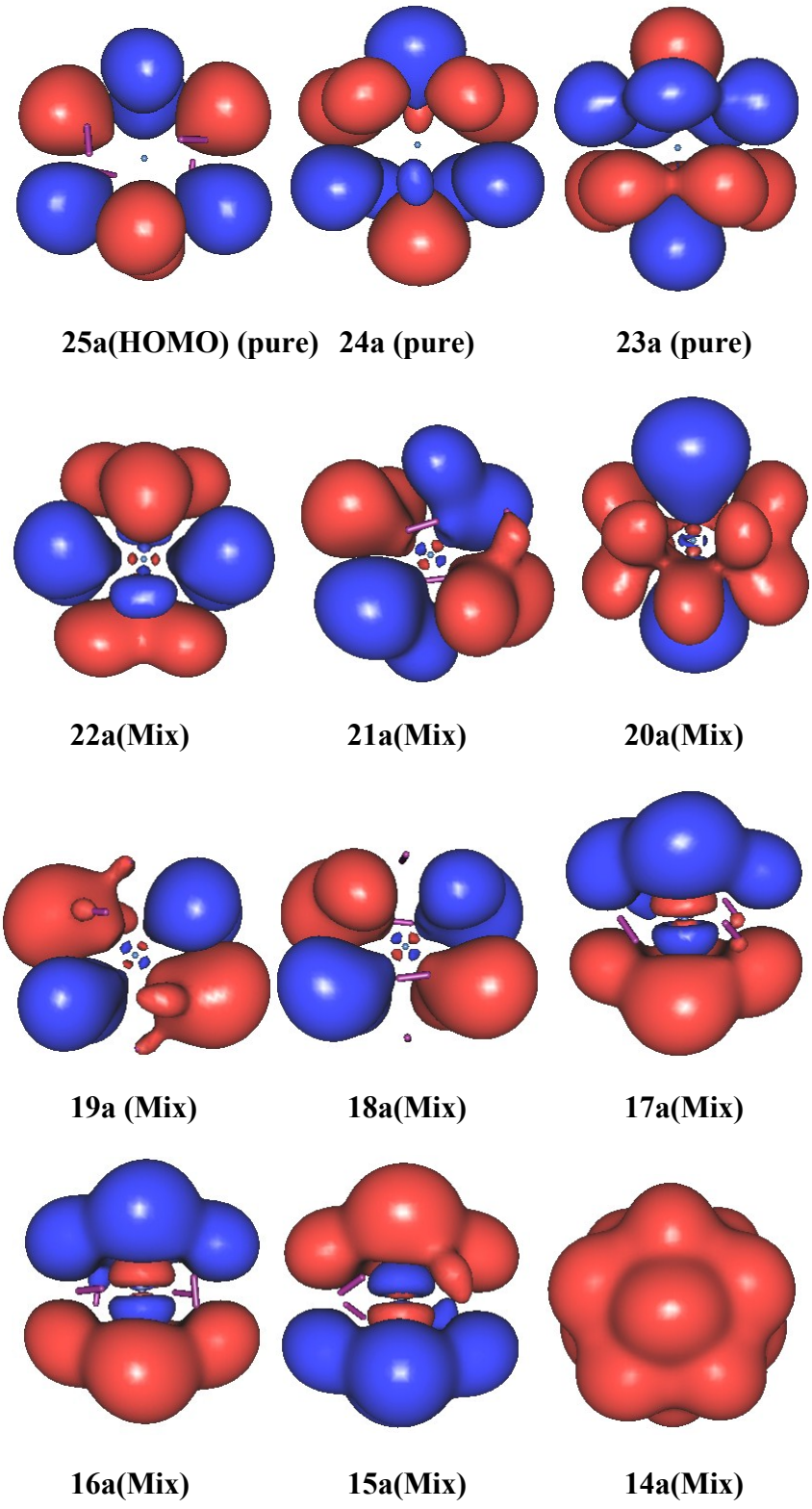


Figure S6. MOs Picture of $Ac(H_2)_{12}^{3+}$ using BHLYP-D3/def-TZVPP method

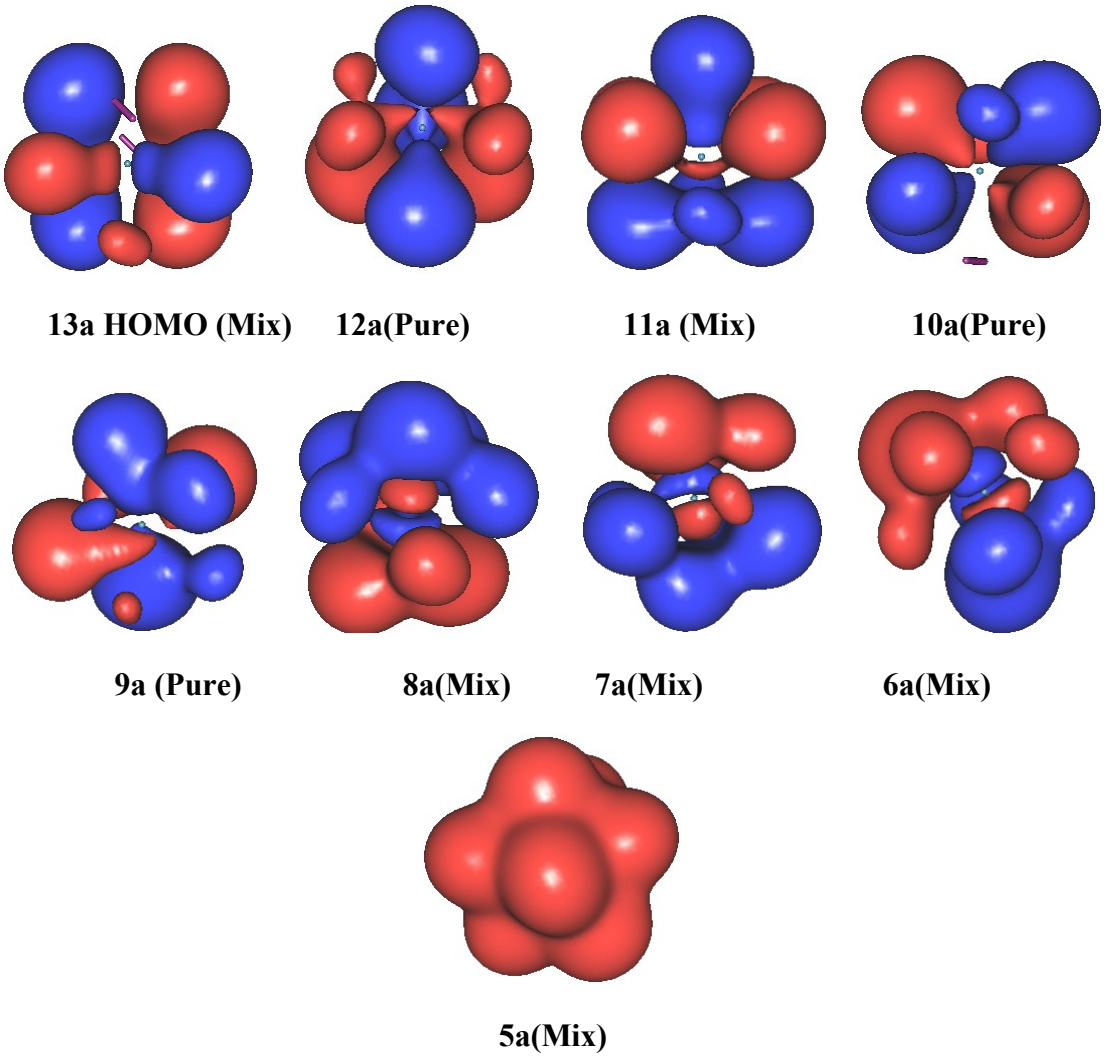
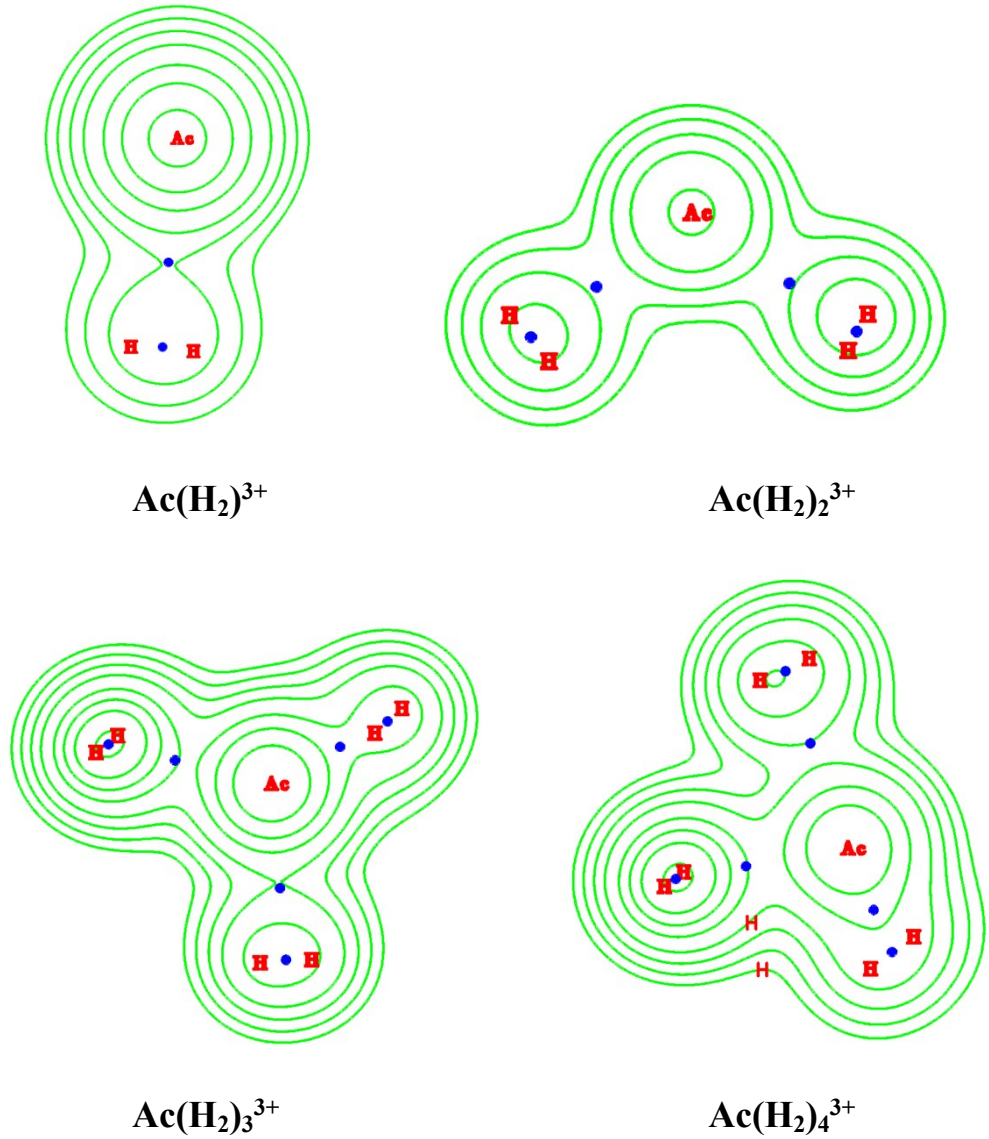


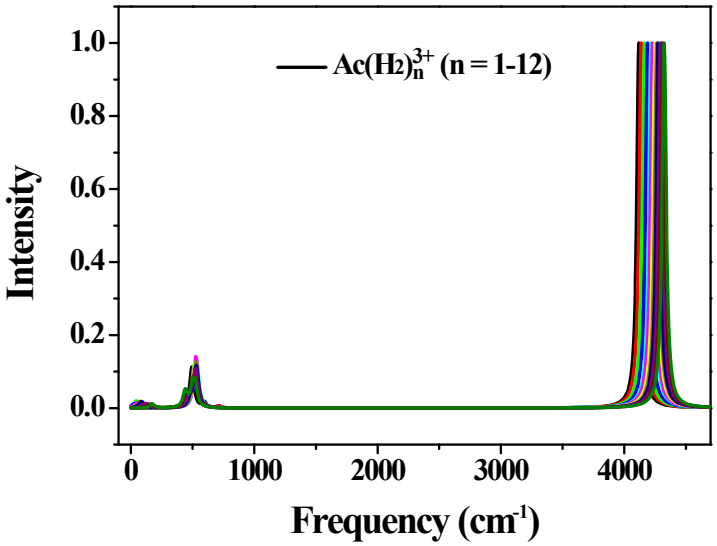
Figure S7. MOs Picture of $La(H_2)_9^{3+}$ using BHLYP-D3/def-TZVPP method



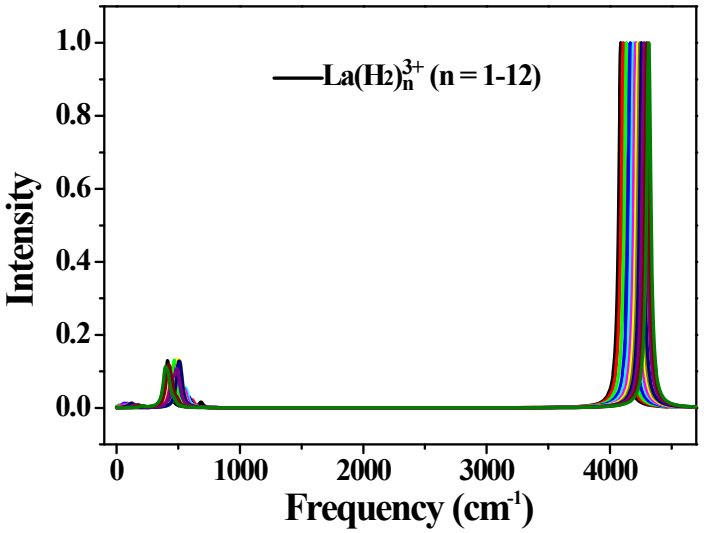
$Ac(H_2)^{3+}$ $Ac(H_2)_2^{3+}$

$Ac(H_2)_3^{3+}$ $Ac(H_2)_4^{3+}$

Figure S8. Electron density plots of $Ac(H_2)_n^{3+}$ (n = 1-4) using BHLYP-D3/def-TZVPP method employed with energy density function.



(a)



(b)

Figure S9. Frequency Plot of a) $Ac(H_2)_n^{3+}$and b) $La(H_2)_n^{3+}$ (n = 1-12) clusters using BHLYP-D3/def-TZVPP method