

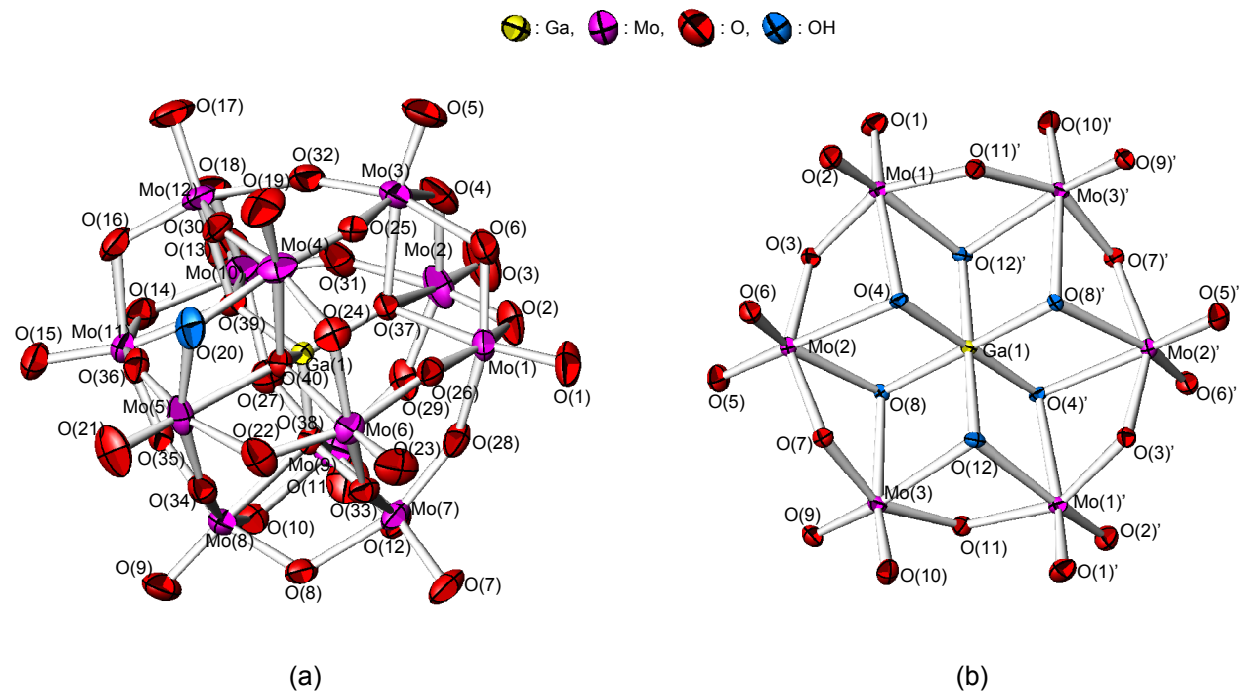


Figure S1. Structures of (a) $[(\text{GaO}_4)\text{Mo}_{12}\text{O}_{35}(\text{OH})]^{4-}$; (b) $[\text{Ga}(\text{OH})_6\text{Mo}_6\text{O}_{18}]^{3-}$.
The thermal ellipsoids are drawn at the 50% probability level.

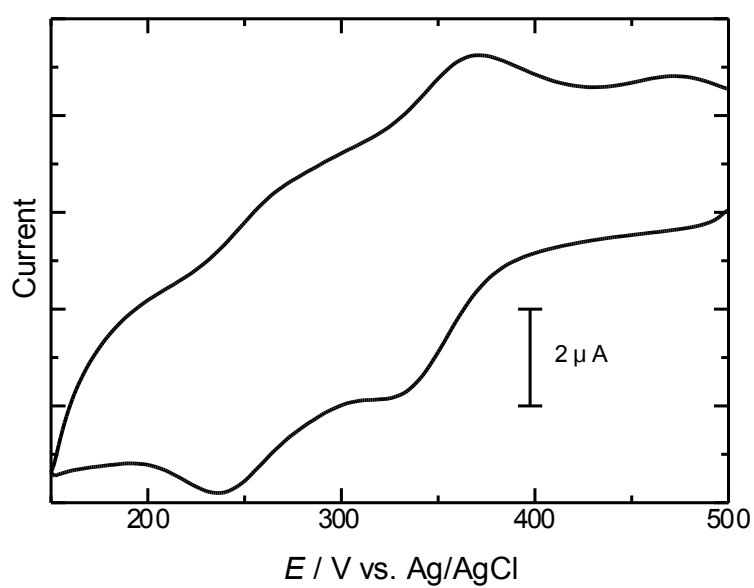


Fig. S2. Cyclic voltammogram of a 5.0 mM Mo^{VI}-0.50 mM Ga^{III}-0.2 M HCl-40% (v/v) CH₃CN system.

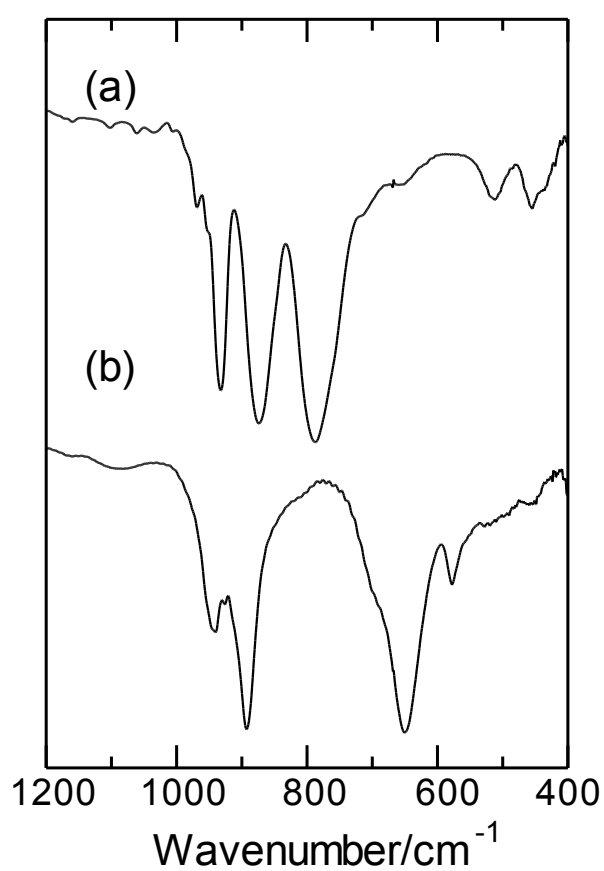


Fig. S3. IR spectra of (a) (NPrⁿ₄)₄[(GaO₄)Mo₁₂O₃₅(OH)]
and (b) K₃[Ga(OH)₆Mo₆O₁₈]·7H₂O in KBr pellets

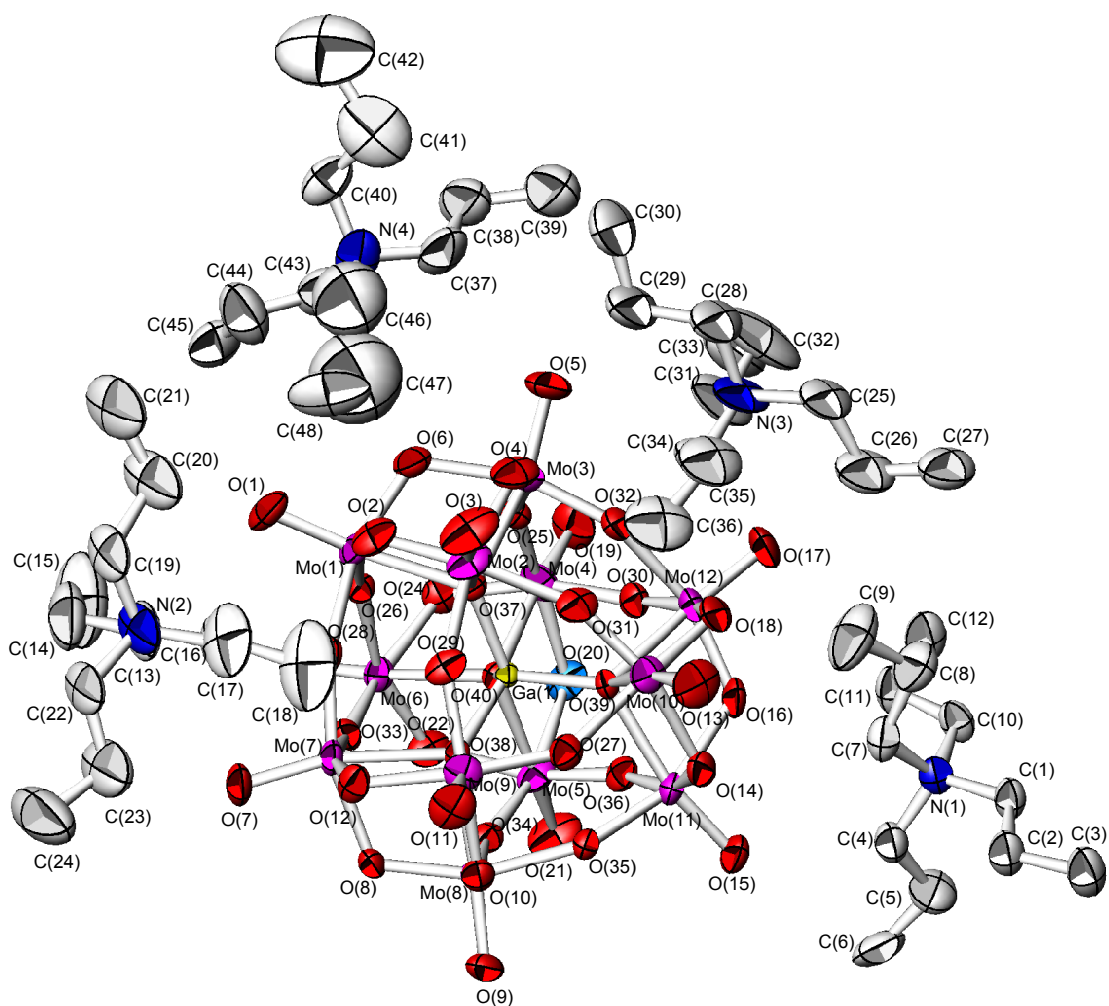


Fig. S4. $\text{NPr}^n_4^+$ cations in $(\text{NPr}^n_4)_4[(\text{GaO}_4)\text{Mo}_{12}\text{O}_{35}(\text{OH})] \cdot n\text{CH}_3\text{CN}$.
The thermal ellipsoids are drawn at the 50% probability level.

Table S1. BVS values of oxygen atoms in $(\text{NPr}^{\text{n}}_4)_4[(\text{GaO}_4)\text{Mo}_{12}\text{O}_{35}(\text{OH})]\cdot n\text{CH}_3\text{CN}$

terminal-O		es- μ_2 -O*		cs- μ_2 -O*		μ_4 -O	
atoms	BVS	atoms	BVS	atoms	BVS	atoms	BVS
O(1)	1.88	O(2)	1.86	O(25)	2.01	O(37)	2.07
O(3)	1.81	O(4)	1.82	O(26)	1.97	O(38)	2.09
O(5)	1.77	O(6)	1.86	O(27)	1.89	O(39)	2.09
O(7)	1.75	O(8)	1.90	O(28)	1.99	O(40)	2.06
O(9)	1.80	O(10)	1.73	O(29)	1.97		
O(11)	1.73	O(12)	1.81	O(30)	1.93		
O(13)	1.84	O(14)	1.87	O(31)	2.02		
O(15)	1.82	O(16)	1.88	O(32)	2.02		
O(17)	1.86	O(18)	1.79	O(33)	1.96		
O(19)	1.79	O(20)	1.41	O(34)	1.97		
O(21)	1.75	O(22)	1.87	O(35)	1.97		
O(27)	1.84	O(24)	1.90	O(36)	1.99		

* es : edge-shared, cs : corner-shared.

Table S2. BVS values of oxygen atoms in $\text{K}_3[\text{Ga}(\text{OH})_6\text{Mo}_6\text{O}_{18}]\cdot 7\text{H}_2\text{O}$

	terminal-O		μ_2 -O		μ_3 -O	
	atoms	BVS	atoms	BVS	atoms	BVS
molecule 1:						
	O(1)	1.71	O(3)	1.89	O(4)	1.23
	O(2)	1.73	O(7)	1.89	O(8)	1.23
	O(5)	1.73	O(11)	1.75	O(12)	1.26
	O(6)	1.75				
	O(9)	1.68				
	O(10)	1.69				
molecule 2:						
	O(13)	1.67	O(15)	1.89	O(16)	1.25
	O(14)	1.72	O(19)	1.88	O(20)	1.19
	O(17)	1.74	O(23)	1.76	O(24)	1.25
	O(18)	1.68				
	O(21)	1.73				
	O(22)	1.66				