Supplementary Information

New Insight into the Structure of $PuGaO_3$ from *ab initio* Particle-swarm Optimization Methodology

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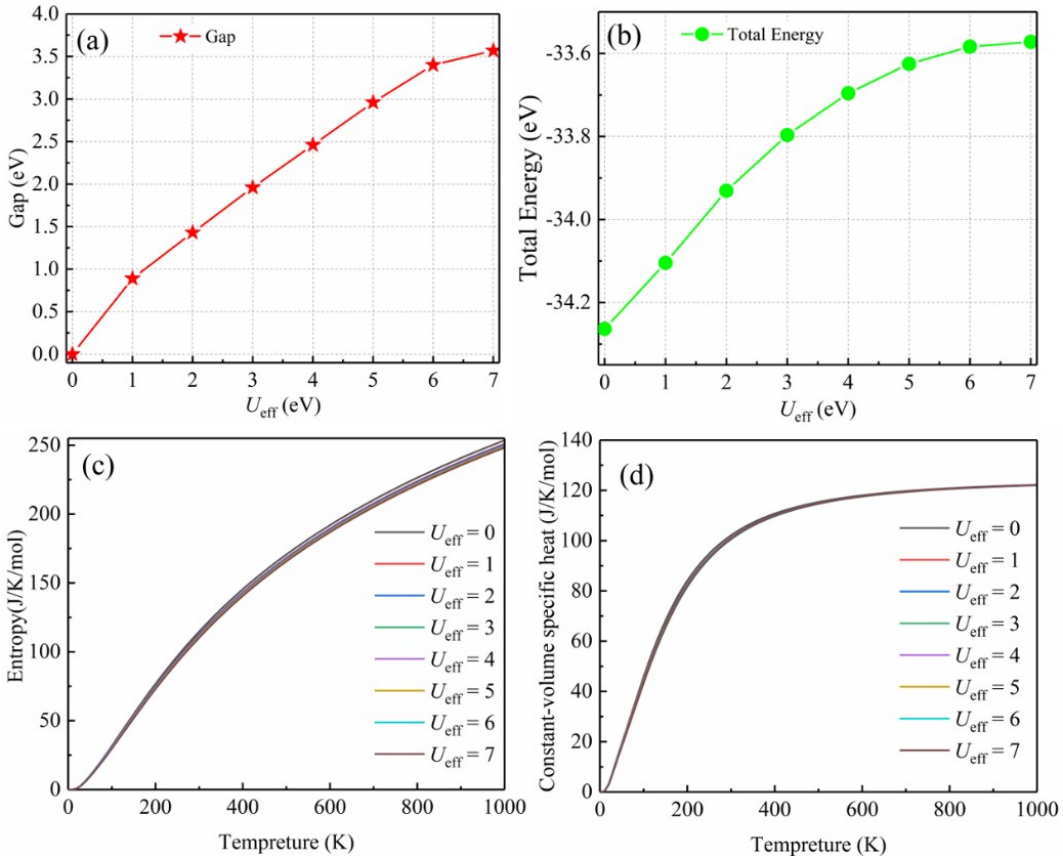

Fig. S1 Dependences of the (a) band gap, (b) total energy, (c) enthalpy and (d) constant-volume specific heat on different U_{eff} for *Pnma* $PuGaO_3$.

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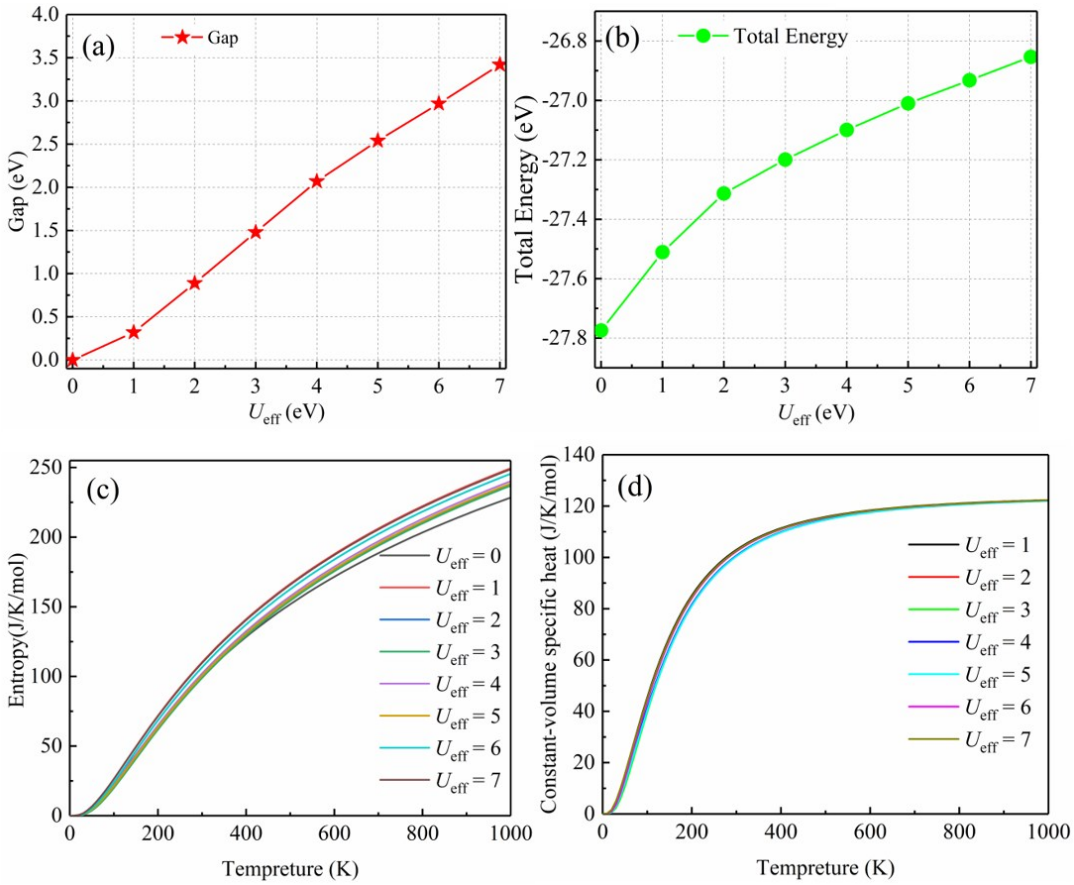

Fig. S2 Dependences of the (a) band gap, (b) total energy, (c) enthalpy and (d) constant-volume specific heat on different U_{eff} for *Pnma* $CeGaO_3$.

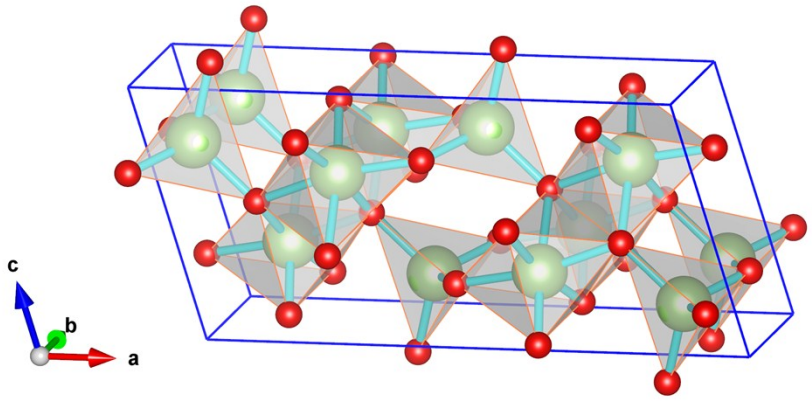

Fig. S3 The monoclinic structure of Ga_2O_3. The lines indicate Ga−O separations shorter than 2.2 Å. Green balls are Ga, red O.

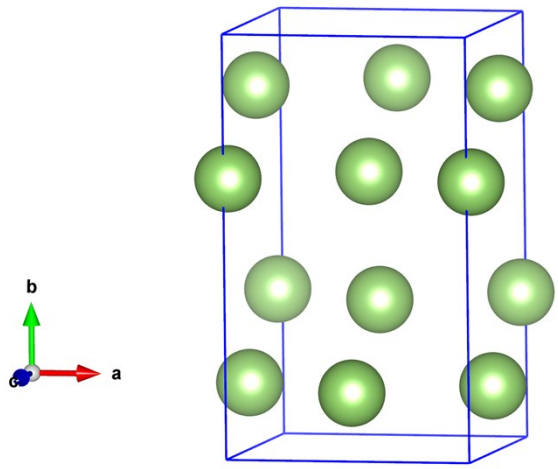

Fig. S4 The orthorhombic structures of Ga.

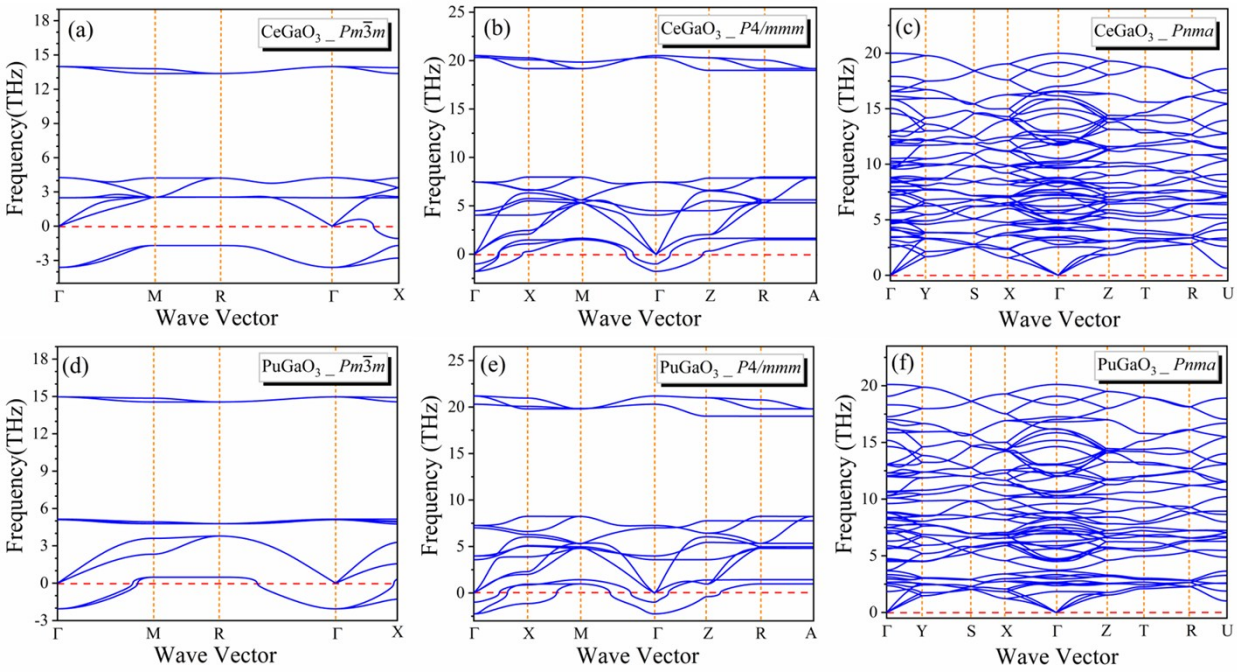

Fig. S5 The calculated phonon dispersion curves of $AGaO_3$ (A = Ce, Pu) in the states of AFM magnetism from PBEsol + U. (a) CeGaO3_ *Pmm*, (b) CeGaO3_*P*4/*mmm*, (c) CeGaO3_*Pnma*, (d) PuGaO3_ *Pmm*, (e) PuGaO3_ *P*4/*mmm*, (f) PuGaO3_*Pnma*.

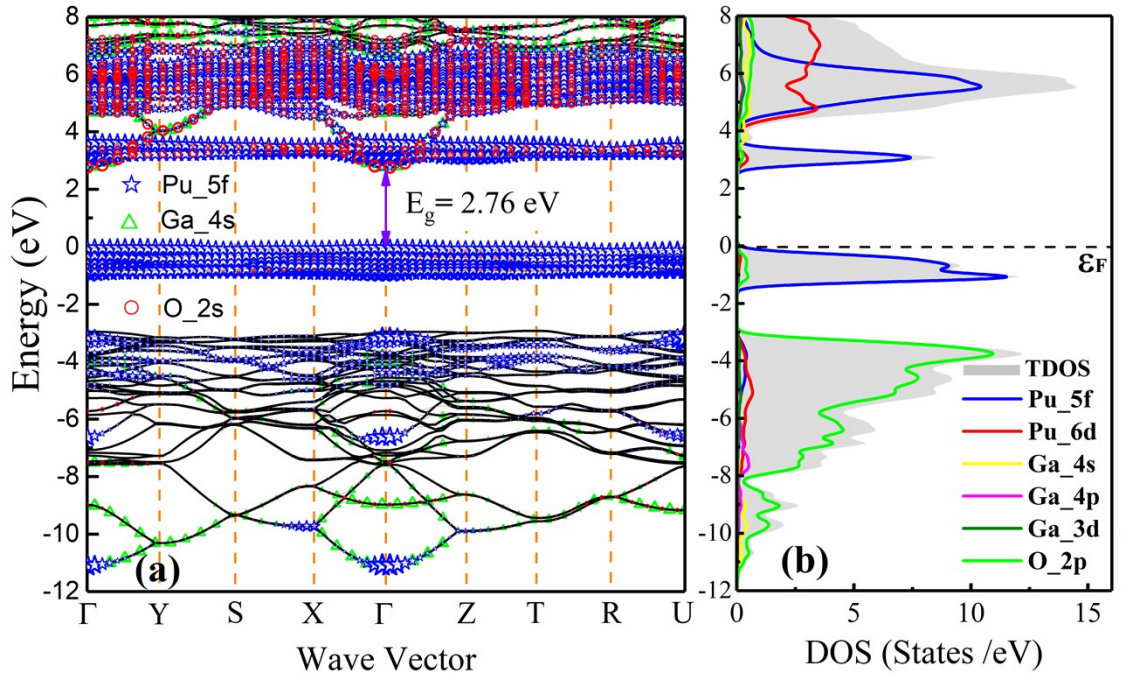

Fig. S6 The calculated (a) fat-band dispersions and (b) PDOS of $PuGaO_3$ for *Pnma* phase in the states of AFM magnetism using HSE06 method. The DOS projected onto Pu-5f, Pu-6d, Ga-4s, Ga-4p, Ga-3d, and O-2p orbitals. Energy is shifted so that the Fermi level ε_F equals to zero.

Supporting Tables

Table S1. Detailed structure information of predicted structures of $PuGaO_3$ and $CeGaO_3$ from PEBsol + *U* calculations.

	Phase	Lattice parameter (Å, deg)	Atom	Wyckoff position	Atomic coordinates (fractional)			Energy (eV/f.u.)
					x	y	z	
$PuGaO_3$	*Pnma*	*a* =5.513	Pu	4c	0.0398	0.2500	0.9931	-33.7028
	(*No*:62, *Z* = 4)	*b* = 7.694	Ga	4b	0.5000	0.0000	0.0000	
		c = 5.467	O_1	8d	0.2926	0. 0402	0.7085	
		$\alpha = \beta = \gamma = 90$	O_2	4c	0.4831	0.2500	0.0782	
$CeGaO_3$	*Pnma*	*a* = 5.508	Ce	4c	0.0349	0.2500	0.9927	-27.1508
	(*No*:62, *Z* = 4)	*b* = 7.794	Ga	4b	0.5000	0.0000	0.0000	
		c = 5.589	O_1	8d	0.2875	0. 0379	0.7136	
		= = = 90	O_2	4c	0.4869	0.2500	0.0715	

Table S2. Calculated lattice constants (a_0, b_0, c_0), the band gap (E_g), and total energy (E) for the *Pnma* $PuGaO_3$ *and* $CeGaO_3$ through PBEsol + U and HSE06 methods [a]

System	V_{xc}	Mothod	a_0 (Å)	b_0 (Å)	c_0 (Å)	EMS	E_g (eV)	E (eV/f.u.)
$PuGaO_3$	PBEsol+U	FM	5.533	7.691	5.477		2.32	-33.696
		AFM	5.513	7.694	5.467		2.46	-33.703
	HSE06	FM	5.465	7.635	5.419		2.51	-49.949
		AFM	5.458	7.637	5.422		2.76	-49.951
$CeGaO_3$	PBEsol+U	FM	5.509	7.795	5.588		2.40	-27.145
		AFM	5.508	7.794	5.589	0.065	2.36	-27.150
	HSE06	FM	5.494	7.716	5.416		2.81	-39.633
		AFM	5.494	7.710	5.418	0.047	2.88	-39.634
		exp.[3]	5.484	7.747	5.490			

[a] $EMS^2 = \frac{1}{3}\left[(a-a_{exp.})^2 + (b-b_{exp.})^2 + (c-c_{exp.})^2\right]$

Table S3. The optical phonon frequencies (unit:cm^{-1}) and LO-TO splitting for infrared active modes at Γ point of $PuGaO_3$. R represents Raman active, IR represents infrared active. The first column (e = 0) is for no electric field, the e//x, e//y and e//z are for the field parallel to x, y and z principal axis, respectively. $\Delta\omega^2 = \Delta\omega_{LO}^2 - \Delta\omega_{TO}^2$

	Modes	TO (e=0)	LO (e//x)	LO (e//y)	LO (e//z)	$\sqrt{\Delta\omega^2}$
Infrared (IR)	B_{1u} (1)	85.893	85.893	85.893	**86.393**	9.285
	B_{1u} (2)	150.504	150.504	150.504	**160.178**	54.821
	B_{1u} (3)	227.524	227.524	227.524	**235.663**	61.399
	B_{1u} (4)	241.834	241.834	241.834	**275.758**	132.509
	B_{1u} (5)	280.761	280.761	280.761	**292.936**	83.575
	B_{1u} (6)	311.749	311.749	311.749	**312.483**	21.403
	B_{1u} (7)	343.705	343.705	343.705	**433.000**	263.355
	B_{1u} (8)	433.000	432.100	432.100	**517.492**	283.388
	B_{1u} (9)	542.376	542.376	542.376	**584.672**	218.334
	B_{2u} (1)	128.522	128.522	**152.172**	128.522	81.476
	B_{2u} (2)	153.773	153.773	**161.011**	153.773	47.734
	B_{2u} (3)	237.264	237.264	**258.746**	237.264	103.223
	B_{2u} (4)	260.781	260.781	**279.527**	260.781	100.642
	B_{2u} (5)	323.524	323.524	**473.595**	323.524	345.867

	B_{2u} (6)	495.810	495.810	**507.685**	495.810	109.162
	B_{2u} (7)	544.544	544.544	**596.313**	544.544	243.025
	B_{3u} (1)	86.260	**86.727**	86.260	86.260	8.988
	B_{3u} (2)	153.940	**161.278**	153.940	153.940	48.096
	B_{3u} (3)	221.620	**227.157**	221.620	221.620	49.849
	B_{3u} (4)	249.439	**256.911**	249.439	249.439	61.509
	B_{3u} (5)	263.016	**287.899**	263.016	263.016	117.085
	B_{3u} (6)	290.368	**389.436**	290.368	290.368	259.514
	B_{3u} (7)	399.043	**454.048**	399.043	399.043	216.620
	B_{3u} (8)	492.308	**515.590**	492.308	492.308	153.188
	B_{3u} (9)	516.891	**574.998**	516.891	516.891	251.886
Raman(R)	A_g (1)	77.787				
	A_g (2)	105.673				
	A_g (3)	197.103				
	A_g (4)	273.923				
	A_g (5)	331.063				
	A_g (6)	353.445				
	A_g (7)	434.001				
	B_{1g} (1)	122.285				
	B_{1g} (2)	188.464				
	B_{1g} (3)	327.961				
	B_{1g} (4)	352.377				
	B_{1g} (5)	578.334				
	B_{2g} (1)	84.425				
	B_{2g} (2)	111.677				
	B_{2g} (3)	205.743				
	B_{2g} (4)	337.167				
	B_{2g} (5)	409.384				
	B_{2g} (6)	488.305				
	B_{2g} (7)	604.319				
	B_{3g} (1)	100.470				
	B_{3g} (2)	293.103				
	B_{3g} (3)	359.349				
	B_{3g} (4)	430.131				
	B_{3g} (5)	635.974				
Silent	A_u (1)	62.443				
	A_u (2)	150.171				
	A_u (3)	176.622				
	A_u (4)	225.189				
	A_u (5)	266.785				

A_u (6)	340.302
A_u (7)	504.483
A_u (8)	527.565