<p style="text-align:center">*Supplementary Information*</p>

Eu³⁺ ions as crystal-field probe for low-symmetry sites in doped phosphors – case study: Eu³⁺ at triclinic sites in Li₆RE(BO₃)₃ (RE = Y, Gd), YBO₃ and ZnO, and at trigonal sites in YAl₃(BO₃)₄

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S1. Crystallographic structure and the axis systems

S1.1. Li₆Y(BO₃)₃

Yttrium lithium double borate, Li₆Y(BO₃)₃, crystallizes in monoclinic system with space group (SG): $P2_1/c$ (no. 14) with four formula units per unit cell ($Z = 4$), and the unit cell parameters are: $a = 7.157$Å, $b = 16.378$Å, and $c = 6.623$Å, and $\beta = 105.32°$.[71] The atomic coordinates were collected from 1501933.cif in the website: Crystallography Open Database.[71c] The Y^{3+} ions occupy triclinic C_1 site and are surrounded by eight oxygen ions in irregular coordination with different Y−O bond distances. The Y−O band distances in YO₈ polyhedron range from 2.2999Å to 2.5197Å and average Y−O bond is 2.376Å. The distorted YO₈ dodecahedra connect each other by sharing edges to form infinite zigzag chains parallel to the c-axis.[38] Eu³⁺ ions are doped at Y^{3+} sites, sine their ionic radii are almost identical: Y^{3+} (1.019Å) and Eu^{3+} (1.066Å).[72] Therefore, we may consider the crystallographic data[71] of undoped Li₆Y(BO₃)₃ crystal as for the doped crystal. Due to the monoclinic crystal symmetry of Li₆Y(BO₃)₃, the CAS is non-Cartesian and hence, a modified CAS* must be chosen for CFP modelling in ECM approach. We adopt the CAS* (a^*, b, c) with Y/Eu at the origin as follows: $X \| a^*$, $Y \| b$, and $Z \| c$, with the a^*-axis perpendicular to the b-c plane. Thus, the a-axis lies in the (a^*c) plane making an angle β away from the c-axis. The atomic coordinates (x, y, z) expressed in CAS may be converted to the CAS* coordinates using relations: $X = x \sin\beta$, $Y = y$ and $Z = x \cos\beta + z$.[34]

S1.2. Li₆Gd(BO₃)₃

Li₆Gd(BO₃)₃ host is isostructural to Li₆Y(BO₃)₃.[37–39] The atomic coordinates and structural parameters are given in.[40] The unit cell parameters are: $a = 7.2277$Å, $b = 16.5075$Å, and $c = 6.6933$Å, and $\beta = 105.3737°$.[40] Since smaller Y atom is replaced by the larger Gd atom,[72] the unit cell volume is larger for Li₆Gd(BO₃)₃ host (V = 769.93 Å³) than for Li₆Y(BO₃)₃ (V = 748.74Å³). The Gd–O bond

distances in GdO_8 polyhedron range from 2.23 Å to 2.597 Å and average Gd–O bond is 2.437Å. Thus, the size of the GdO_8 polyhedron is also larger than that of YO_8 polyhedron in $Li_6Y(BO_3)_3$.

S1.3. YBO₃

Yttrium orthoborate, YBO_3, crystal belongs to monoclinic structure with SG: $C2/c$ (no. 15) and Z =12. The unit cell parameters are: a = 11.314 Å, b = 6.5404 Å, c = 9.5503 Å, and β = 112.902°.[41] The Y^{3+} ions are coordinated to eight oxygens, but two bridging oxygens are quite distant. Hence, the Y^{3+} ions occupy two distinct Wyckoff sites, 4c and 8f, with triclinic site symmetry, C_i and C_1, respectively. The ratio of Y^{3+} (C_1) sites to Y^{3+} (C_i) sites is 2:1.[41] Eu^{3+} ions replace Y^{3+} ions in YBO_3:Eu^{3+}.[15] Average Y/Eu−O bond distance is 2.384 Å at the C_i site, while 2.356 Å at the C_1 site.[41] The CAS* (a*, b, c) were also adopted for CFP modelling of YBO_3:Eu^{3+} as for $Li_6RE(BO_3)_3$.

S1.4. ZnO

ZnO crystallizes in wurtzite nanostructure having hexagonal SG: $P6_3mc$ - C_{6v}^4 (no. 186).[17,18] The unit cell parameters are: a = b = 3.28Å, c = 5.17 Å, γ = 120° and Z = 2. Zn^{2+} occupies a trigonal geometry with C_{3v} point symmetry and is coordinated to four oxygens in pure ZnO. There are two sets of Zn-O bonds in Zn-O_4 tetrahedron: one Zn−O(2) bond of length 1.9853Å and three Zn-O(1) bonds of length 1.9864Å. The bond angle O(2)−Zn−O(1) is 107.8325°, while O(1)−Zn−O(1) angles are 111.06°. Zn^{2+} is replaced by Eu^{3+} ions in ZnO:Eu^{3+} nanocrystals. The mismatch of ionic radius and charge compensation of host-dopant (Zn^{2+} -Eu^{3+}) ions induce significant lattice distortion or strain, and thus, lowering of the actual trigonal C_{3v} symmetry at substitutional sites.[16,17] Since four coordination is too low for RE^{3+} ions,[19] there are two distinct sites for Eu^{3+} substitutions in ZnO: surface site (denoted as the A-sites) having C_1 symmetry and lattice sites (B-sites) having C_s or C_1 symmetry. A- and B- sites are coordinated to seven and four oxygen ions, respectively.[16] Since the hexagonal axes do not form any Cartesian axis system, for CFP modelling we adopted a CAS* with Zn/Eu at the origin, as follows:[34] the Z axis lies along the crystal c-axis (which is parallel to the C_3 axis),[19] the Y axis bisects the angle γ, the X axis being normal to the YZ plane in the right-handed sense, i.e., $X = \sqrt{3}(x - y)/2$, $Y = (x + y)/2$, $Z = z$.

S1.5. YAl₃(BO₃)₄

Yttrium aluminium borate, $YAl_3(BO_3)_4$, crystallizes in hexagonal structure having SG: $R32$ (no. 155). The unit cell parameters are: a = b = 9.295Å, c = 7.243 Å, γ = 120° and Z = 3.[73] Y^{3+} is replaced by Eu^{3+} ions and is coordinated to six oxygens. Y/Eu^{3+} occupies a trigonal geometry with D_3 point symmetry. The Eu^{3+} ions are located on the three-fold rotary axis C_3 in slightly distorted prisms in which the upper and lower triangles are slightly rotated relative to each other by an angle 2Φ.[74] All six Y/Eu−O3 bond lengths are equal to 2.154 Å, while O3−Y/Eu−O3 angles, *i.e.*, 142.155°, 114.714°,

98.17°, 60.532°, differ substantially from the characteristic octahedral angles of 180° and 90°, giving rise to the trigonal distortion of Y/Eu-O_6 polyhedra in $YAl_3(BO_3)_4$. The CAS* for CFP modelling is the same as for the hexagonal $ZnO:Eu^{3+}$ crystal.

S2. Review of experimental energy levels and CF calculations

S2.1. $Li_6Y(BO_3)_3:Eu^{3+}$

Photoluminescence spectra of Eu^{3+} doped yttrium lithium double borates, $Li_6Y(BO_3)_3$, were recorded at 77 K by Hölsä and Leskelä[38] and also at 300K by Ju *et al.*[39] The experimentally determined 24 energy levels including 3 for $^7F_1$, 5 for $^7F_2$, 6 for $^7F_3$, 8 for $^7F_4$ due to $^5D_0 \rightarrow {}^7F_{0-4}$ transitions in the energy range up to 17250 cm^{-1} were reported by Hölsä and Leskelä.[38] The observed barycentre of the energy bands was estimated at: 0 ($^7F_0$), 357 cm^{-1} ($^7F_1$), 900 cm^{-1} ($^7F_2$), 1898 cm^{-1} ($^7F_3$), 3011 cm^{-1} ($^7F_4$) and 17234 cm^{-1} ($^5D_0$).[38] The overall CF splittings of the $^7F_{1-4}$ multiplets are, respectively: 144, 130, 198, 368 (all in cm^{-1}). The E_i were calculated using an approximated CF Hamiltonian as a function of only two CFPs, *e.g.*, B_{20} and B_{22}. However, calculated E_i were not listed therein.[38] The fitted CFPs were grossly inadequate and incompatible with the fact that no point symmetry exists for which CF Hamiltonian can be parametrized by two CFPs only. Furthermore, for the low triclinic symmetry, several possible sets of the *non-standard* CFPs can be obtained in fittings of the observed energy levels for 4f ions.[23,34,38] For example, three sets of B_{20} and B_{22} were obtained for $Li_6RE(BO_3)_3:Eu^{3+}$ (RE = Y, Gd).[38] Thus, since these CFP sets are physically equivalent but numerically distinct, they cannot be related to any particular axis system.[23,27] Alternatively, no axis system can be assigned to the CFP sets fitted for $Li_6Y(BO_3)_3:Eu^{3+}$.[38] This realization has led to introduction of the notion of a *nominal* axis system,[23] *i.e.*, an undefined Cartesian axis system assigned to a given alternative CFP set. The *nominal* axes cannot be *a priori* related to any well-defined axes in crystal. Orientation of a particular *nominal* axis system, in which a given alternative CFP set is expressed, may be established by comparison with the modelled CFPs calculated using, *e.g.*, ECM[49,50] or SPM[51-53] in a well-defined axis system,[53] *e.g.*, symmetry adapted axis system (SAAS), CAS or CAS*.[21–27,37]

The photoluminescence spectra of $Li_6Y(BO_3)_3:Eu^{3+}$ at room temperature exhibited some sharp peaks in the visible and UV region.[39] The CF splitting components were not observed in[39] due to poor resolution of the spectrophotometer used. The sharp peaks were ascribed to the intra-configurational 4f - 4f transitions (absorption) of Eu^{3+} in the host $Li_6Y(BO_3)_3$ crystal: $^7F_0 \rightarrow {}^5F_4$, $^5H_6$, $^5H_3$, $^5D_4$, (^5G_J and $^5L_7$ manifold), $^5L_6$, $^5D_3$, $^5D_2$, $^5D_1$ at energies: 33557, 31446, 30675, 27701, 26455, 25445, 23981, 21552, 19048 (all in cm^{-1}), respectively. In addition, the sample showed groups of sharp peaks in the emission spectra due to $^5D_0 \rightarrow {}^7F_{0-4}$ transitions.[39] The E_i from the emission spectra were assigned as follows (all in cm^{-1}): 0 ($^7F_0$), 405 ($^7F_1$), 925 ($^7F_2$), 1803 ($^7F_3$), 3067 ($^7F_4$), 17212 ($^5D_0$), 19048 ($^5D_1$),

21552 ($^5D_2$), 23981 ($^5D_3$), 27701 ($^5D_4$), 25445 ($^5L_6$), 26455 ($^5L_7$), 30675 ($^5H_3$), 31446 ($^5H_6$) and 33557 ($^5F_4$). However, calculations of E_i were not performed in [39].

S2.2. Li$_6$Gd(BO$_3$)$_3$:Eu^{3+}

Photoluminescence spectra of Eu^{3+} doped in Li$_6$Gd(BO$_3$)$_3$ were recorded at 77 K by Hölsä and Leskelä.[38] The observed barycentre of the energy bands is found at (all in cm^{-1}): 0 ($^7F_0$), 356 ($^7F_1$), 900 ($^7F_2$), 1898 ($^7F_3$), 2999 ($^7F_4$) and 17238 ($^5D_0$).[38] The comparison of the observed barycentre of the $^{2S+1}L_J$ multiplets of Eu^{3+} ions in different hosts, *i.e.*, Li$_6$Y(BO$_3$)$_3$ and Li$_6$Gd(BO$_3$)$_3$, reveals only slight variations which may be due to the nephelauxetic effect.[19,38] The E_i were also calculated[38] using four different sets of only two CFPs, *i.e.*, B_{20} and B_{22}, though calculated levels were not provided by Hölsä and Leskelä.[38] The CF splittings of $^7F_{1-4}$ multiplets are larger for the Li$_6$Gd(BO$_3$)$_3$ host (162, 122, 207, 372 cm^{-1}) than for Li$_6$Y(BO$_3$)$_3$ host[38] and hence the matched CFPs are somewhat larger for Li$_6$Gd(BO$_3$)$_3$ host than for Li$_6$Y(BO$_3$)$_3$.[38]

S2.3. YBO$_3$:Eu^{3+}

Photoluminescence spectra of YBO$_3$:Eu^{3+} (0.5 atom%) crystal was measured at 10 K.[15] The samples did not exhibit any characteristics of phase transitions on cooling to 10 K, so that the Eu^{3+} site symmetry can be taken as in pure crystal.[15] The emission and absorption spectra exhibited characteristic features of two distinct Eu^{3+} sites of triclinic symmetry without inversion (C_1) and with inversion (C_i). Total 31 levels were experimentally assigned for Eu^{3+} ions in the C_1 sites, including 3 for $^7F_1$, 5 for $^7F_2$, 6 for $^7F_3$, 7 for $^7F_4$ due to $^5D_0 \rightarrow {^7F_{0-4}}$ emissions in the energy range below 17200 cm^{-1}, as well as 3 for $^5D_1$ and 5 for $^5D_2$ level due to $^7F_0 \rightarrow {^5D_{1,2}}$ absorption within the energy range of 17200 – 21470 cm^{-1}, as consistent with the selection rules for the C_1 site. On the other hand, only 7 energy levels, including 3 for $^7F_1$ and 3 for $^5D_1$ were detected in the emission and absorption spectra for Eu^{3+} ions at the C_i symmetry.[15] As expected, the transition $^5D_0 \rightarrow {^7F_0}$ was not observed as expected for Eu^{3+} ion at the C_i site.[15] Since only few E_i for C_i symmetry were detected in luminescence spectra, the energy level calculations[15] was mostly limited to the levels of C_1 site. Instead of taking the actual C_1 symmetry at Eu^{3+} ion site, Jia *et al.*[15] employed ascend/descend symmetry (ADS) method[19,22,27,36] in the chain: 'triclinic (C_1, C_i) $\rightarrow$ orthorhombic (C_{2v}) $\rightarrow$ cubic (O_h)' to minimize the number (total 27) of CFPs required for the energy level calculations. Thus, in the spirit of ADS, only nine CFPs, which are all real, were taken in the Hamiltonian: cubic CFPs ($B_4{}^C$, $B_6{}^C$), orthorhombic CFPs (B_{20}, B_{22}, B_{42}, B_{44}, B_{62}, B_{64} and B_{66}).[15] All imaginary CFPs (in Wybourne notation)[19] were set to zero in this approximation.[15] Further, only six non-zero values of CFPs, with large standard deviation of the parameters, were reported, and three CFPs, *i.e.*, B_{62}, B_{64} and B_{66}, were constrained to zero. From the so-obtained CFP values, it is evident that these approximated symmetry cases correspond to different orientation of the axis system w.r.t. the CAS or the CAS* adopted above for Eu^{3+} at triclinic site. Therefore, it is essential to define precisely the axis system in which

particular higher symmetry approximation is achieved in order to enable CFPs modelling as well as to validate the applicability of the ADS method for analysis of the observed E_i. Furthermore, if the ADS method is applied, the assignment of a well-defined axis system to a given higher symmetry approximation is necessary to enable comparison of CFPs resulting in the actual triclinic symmetry.[27] The axis system was not explicitly defined by Jia *et al.*[15] for the actual triclinic symmetry case or for the assumed higher symmetry cases for YBO_3:Eu^{3+}.

S2.4. ZnO:Eu^{3+}

Optical spectra of Eu^{3+} doped ZnO nanocrystals were recorded at 10 K.[16] The surface site (A) exhibits inhomogeneous broadening of excitation and emission spectra of Eu^{3+} due to a very distorted local environment. The lattice site (B) shows very sharp and intense spectra due to the bulk crystalline surrounding of Eu^{3+} ions. Very sharp f-f transitions, $^5D_0 \rightarrow {}^7F_J$ ($J = 0 - 4$) and weak emission lines corresponding to $^5D_1 \rightarrow {}^7F_J$ ($J = 1 - 4$) were detected at 10 K. Three lines for $^5D_0 \rightarrow {}^7F_1$ transitions and five lines for $^5D_0 \rightarrow {}^7F_2$ transitions of Eu^{3+} were observed, suggesting descend of symmetry from actual trigonal C_{3v} to triclinic C_1 symmetry for the A-site and to C_s or C_1 (monoclinic/triclinic) symmetry for the B-site. A total number of 12 energy levels for A-site and 25 energy levels for B-site were identified.[16] The width of the $^7F_1$ level for Eu^{3+} ions at the A-sites (= 147 cm^{-1}) is larger than that at the B-sites (= 134 cm^{-1}), implying relatively stronger CF strength for the A-site due to higher oxygen coordination. However, no CF calculations using the observed spectra were performed in Ref. 16.

S2.5. YAl$_3$(BO$_3$)$_4$

Fluorescence, absorption and magnetic circular dichroism (MCD) spectra of $YAl_3(BO_3)_4$:Eu^{3+} were recorded at 77 K and 4.2 K by Görller-Warland *et al.*[11] Later, luminescence emission and absorption spectra of $YAl_3(BO_3)_4$:Eu^{3+} were also recorded at room temperature by Ben Amar *et al.*[12] The emission and absorption spectra arise due to transitions between the states arising from different $^{2S+1}L_J$ multiplets of Eu^{3+} ions in the trigonal D_3 site symmetry.[12] The absorption spectrum originates from the $^7F_{0,1,2} \rightarrow {}^5D_{0-3}$ transitions within the energy range in the 15,000–30,000 cm^{-1}, whereas the emission spectrum of $YAl_3(BO_3)_4$:Eu^{3+} corresponds to the $^5D_0 \rightarrow {}^7F_J$ transitions in the range 13000 – 20000 cm^{-1}. The energy level scheme associated with the $^{2S+1}L_J$ multiplets of Eu^{3+} ion within the energy range was determined as:[11] $0 - 5353$ cm^{-1} ($^7F_{0-6}$), 17215 cm^{-1} ($^5D_3$), 18965 -24620 cm^{-1} ($^5D_{0-3}$) and 24877$-$ 25341 cm^{-1} ($^5L_6$). The CFP modelling and calculations of E_i were performed using two different approaches:[11] lattice sum calculations method, and ADS method applying to symmetry lowering from approximated hexagonal $D_{3h} \rightarrow$ actual trigonal D_3 site symmetry,[11] but the axis system used for the CFPs modelling was not defined therein.[11] Using the transition energies, Ben Amor *et al.*[12] also listed the observed E_i (all in cm^{-1}): $0 - 4450$ ($^7F_{0-5}$), 17250$-$ 24400 ($^5D_{0-3}$), 25250$-$ 26050

($^5L_{6,7}$), 26200– 26600 ($^5G_{2,4}$) and at 27642 ($^5D_4$). The E_i were calculated using a model Hamiltonian consisting of the free-ion terms only, disregarding the CF terms for the D_3 symmetry of Eu^{3+} site.[12]

Table S1 Survey of free-ion parameters (FIPs) (in cm^{-1}) for Eu^{3+} ions in respective systems (in cm^{-1}): (b) mean value of FIPs of Eu^{3+} ions in different hosts, taken from Table 5;[19] (c) and (d) for two $Li_6Y(BO_3)_3:Eu^{3+}$ samples studied in [38] and [39], respectively; (e) for $Li_6Gd(BO_3)_3:Eu^{3+}$;[38] (f) C_1 symmetry site and (g) C_i symmetry site for $YBO_3:Eu^{3+}$;[15] (h) A-site and (i) B-site for $ZnO:Eu^{3+}$;[16] (j) for $YAl_3(BO_3)_4:Eu^{3+}$.[11] In columns (c) − (j), only the varied FIPs are listed, while other FIPs are kept constant at the values as in (b). For definitions of FIPs, see, text

FIPs		$Li_6Y(BO_3)_3$		$Li_6Gd(BO_3)_3$	YBO_3		ZnO		$YAl_3(BO_3)_4$
(a)	(b)	(c)	(d)	(e)	(f)	(g)	(h)	(i)	(j)
E_{AVE}	63736								
F^2	82786	82664	82506	82478	82481	82658	82694	82718	82744
F^4	59401	59125	59248	59462	59202	58939	58998	59123	59371
F^6	42644	42680	42702	42809	42464	42551	42432	42487	42612
α	19.80								
β	−617								
γ	1460								
T^2	370								
T^3	40								
T^4	40								
T^6	−330								
T^7	380								
T^8	370								
ζ_{4f}	1332	1323	1325	1324	1326	1331	1327	1333	1330
M^0	2.38								
M^2	1.33								
M^4	0.90								
P^2	303								
P^4	227								
P^6	152								

Table S2 $Li_6Y(BO_3)_3:Eu^{3+}$: (a) Allowed crystal-field parameters (CFPs) for the triclinic C_1 point symmetry at Eu^{3+} site; the calculated CFPs values (cm^{-1}) employing: (b) the ECM method and (c) SPM method along with the rotational invariants, S_k; triclinic ECM/CFPs and SPM/CFPs standardized using the package: 3DD - (b') and (c'); (d) $^{2S+1}L_J$ multiplets of Eu^{3+} ion; (e) experimental[38] and (f) calculated energy (cm^{-1}) levels; (g) the largest and second largest M_J values of the $|^{2S+1}L_J, M_J\rangle$ levels and (h) varied values of ECM (SPM)/CFPs are tabulated

| CFPs | | | | | Multiplet | $E_{\exp}$ | E_i^{cal} | M_J | Varied ECM (SPM)/CFPs |
| | ECM | SPM | 3DD* | | | | | | |
(a)	(b)	(c)	(b′)	(c′)	(d)	(e)	(f)	(g)	(h)
B_{20}	66	113	−397	409	$^7F_0$	0	0	0	
B_{21}^r	214	−164	0	0	$^7F_1$	269	267	−1, 1	$B_{21}^r = 343\ (−312)$
B_{21}^i	−183	178	0	0		386	383	1, 0	
B_{22}^r	111	155	−138	119		413	416	0, 1	$B_{21}^r = 137\ (−98)$
B_{22}^i	64	−93	0	0	$^7F_2$	837	837	0, 2	$B_{44}^r = 330\ (294)$
B_{40}	1128	1142	−137	−749		856	862	−2, 2	
B_{41}^r	563	567	−223	132		908	910	2, −2	
B_{41}^i	178	164	−171	−352		926	925	−1, 1	$B_{20} = 120\ (181)$
B_{42}^r	−244	−232	−429	516		967	970	1, −1	$B_{40} = 1308\ (1278)$
B_{42}^i	67	71	69	−274	$^7F_3$	1800	1808	−2, 2	$B_{66}^r = 354\ (368)$
B_{43}^r	312	278	−705	70		1830	1827	2, −2	
B_{43}^i	24	31	−649	525		1849	1848	−3, 3	
B_{44}^r	251	270	47	153		-	1871	3, −3	
B_{44}^i	−52	−48	−122	−381		1934	1928	1, −1	$B_{66}^r = 202\ (204)$
B_{60}	140	142	171	13		1977	1972	0, −1	
B_{61}^r	157	155	−151	−44		1998	1993	0, −1	
B_{61}^i	13	15	−87	132	$^7F_4$	2815	2815	−3, 3	$B_{62}^r = 308\ (314)$
B_{62}^r	220	243	114	−173		2884	2877	−3, 3	$B_{66}^r = 385\ (391)$
B_{62}^i	−2	−3	92	144		-	2895	2, −2	
B_{63}^r	−55	−59	208	173		2912	2917	−2, 2	
B_{63}^i	17	20	107	136		2994	3002	0, −2	
B_{64}^r	7	3	−30	105		3042	3048	−1, 4	
B_{64}^i	−78	−20	−154	−86		3086	3092	1, 0	
B_{65}^r	−134	−134	27	199		3168	3162	−4, 0	
B_{65}^i	10	−54	108	92		3183	3180	0, −4	

B_{66}^r	276	265	−23	−71	$^5D_0$	17234	17237	0	
B_{66}^i	−20	−52	169	−36					
S_2	198	197	198	198					
S_4	519	519	519	519					
S_6	170	172	170	172					

* The rhombicity ratio: $\kappa = B_{22}^r/B_{20}$ is: 0.35 (b′), 0.29 (c′). The Euler angles are: α = 59.74, β = 54.89, γ = 62.41 (b′); α = 27.93, β = 46.60, γ = 145.67 (c′) (all in degree).

Table S3 Li$_6$Y(BO$_3$)$_3$:Eu^{3+}: (a) Allowed crystal-field parameters (CFPs) for the triclinic C_1 point symmetry at Eu^{3+} site; the calculated CFPs values (cm^{-1}) employing: (b) the ECM method and (c) the SPM method along with the rotational invariants, S_k; triclinic ECM/CFPs and SPM/CFPs standardized using the package: 3DD - (b′) and (c′); (d) $^{2S+1}L_J$ multiplets of Eu^{3+} ion; (e) experimental[39] and (f) calculated energy (cm^{-1}) levels; (g) the calculated barycenter are tabulated

CFPs					Multiplet	E_{exp}	E_i^{cal}	E^{avg}
	ECM	SPM	3DD*					
	(b)	(c)	(b′)	(c′)	(d)	(e)	(f)	(g)
(a)								
B_{20}	71	120	−419	431	$^7F_0$	0	0	0
B_{21}^r	223	−174	0	0	$^7F_1$	405	318, 410, 478	402
B_{21}^i	−192	188	0	0	$^7F_2$	925	804, 854, 862, 1052, 1086	931
B_{22}^r	120	162	−143	124	$^7F_3$	1803	1749, 1757, 1773, 1781, 1837, 1853, 1856	1801
B_{22}^i	70	−97	0	0				
B_{40}	1291	1302	−155	−858	$^7F_4$	3067	2798, 2936, 2968, 3048, 3103, 3147, 3163, 3188, 3208	3062
B_{41}^r	644	647	−247	138				
B_{41}^i	203	187	−201	−399				
B_{42}^r	−275	−264	−470	586				
B_{42}^i	76	81	87	−324	$^5D_0$	17211	17210	17210
B_{43}^r	347	317	−827	90	$^5D_1$	19048	19018, 19044, 19102	19054
B_{43}^i	31	35	−726	597	$^5D_2$	21552	21512, 21528, 21544, 21568, 21584	21547

B_{44}^r	288	308	40	168				
B_{44}^i	−61	−54	−158	−435	$^5D_3$	23981	23956, 23959, 23969, 23978, 23983, 24003, 24007	23980
B_{60}	174	173	230	18				
B_{61}^r	194	188	−184	−49				
B_{61}^i	16	18	−106	159	$^5L_6$	25445	25200, 25204, 25240, 25267, 25287, 25312, 25460, 25530, 25604, 25659, 25668, 25671, 25674	25444
B_{62}^r	276	295	130	−210				
B_{62}^i	−1	−3	103	176				
B_{63}^r	−68	−72	259	211				
B_{63}^i	20	24	139	166				
B_{64}^r	6	3	−37	127				
B_{64}^i	−91	−25	−183	−107				
B_{65}^r	−164	−164	37	245				
B_{65}^i	12	−65	126	106				
B_{66}^r	337	322	−19	−87				
B_{66}^i	−23	−63	214	−40				
S_2	208	208	208	208				
S_4	592	592	592	592				
S_6	208	209	209	208				

* The rhombicity ratio: $\kappa = B_{22}^r/B_{20}$ is: 0.34 (b′), 0.29 (c′). The Euler angles are: α = 60.21, β = 55.28, γ = 61.75 (b′); α = 28.02, β = 46.42, γ = 145.33 (c′) (all in degree).

Table S4 The spherical polar coordinates (R_h, θ_h, ϕ_h) for the O^{2-} ligands around the metal center in the distorted YO_8 polyhedron in $Li_6Y(BO_3)_3$ [71] and $Li_6Gd(BO_3)_3$ [40] host crystal obtained in the CAS* ($a*//X$, $b//Y$, $c//Z$). For explanations, see, text

O^{2-} ion	$Li_6Y(BO_3)_3$			$Li_6Gd(BO_3)_3$		
	R_h (Å)	θ_h (°)	ϕ_h (°)	R_h (Å)	θ_h (°)	ϕ_h (°)
O2	2.359	86.447	−3.066	2.425	84.087	−8.144
O3	2.420	87.061	55.515	2.526	85.143	55.380
O5	2.356	90.291	−174.860	2.516	93.413	−174.533
O6	2.384	99.048	126.462	2.532	100.080	130.118
O8	2.354	63.229	−77.252	2.274	59.052	−76.086
O8′	2.300	168.151	10.814	2.231	167.373	−15.910
O9	2.317	14.008	151.362	2.395	18.016	161.687
O9′	2.520	114.973	−102.443	2.597	114.310	−107.286

Table S5 $Li_6Gd(BO_3)_3$:Eu^{3+}: (a) Allowed crystal-field parameters (CFPs) for the triclinic C_1 point symmetry at Eu^{3+} site; the calculated CFPs values (cm^{-1}) employing: (b) the ECM method and (c) the SPM method along with the rotational invariants, S_k; triclinic ECM/CFPs and SPM/CFPs standardized using the package 3DD- (b′) and (c′); (d) $^{2S+1}L_J$ multiplets of Eu^{3+} ion; (e) experimental[38] and (f) calculated energy (cm^{-1}) levels; (g) the largest and second largest M_J values of the $|^{2S+1}L_J, M_J\rangle$ levels; and (h) varied values of ECM (SPM)/CFPs are tabulated

CFPs					Multiplet	E_{exp}	E_i^{cal}	M_J	Varied ECM (SPM)/ CFPs
	ECM	SPM	3DD*						
(a)	(b)	(c)	(b′)	(c′)	(d)	(e)	(f)	(g)	(h)
B_{20}	740	755	−1132	−1060	$^7F_0$	0	0	0	
B_{21}^r	148	242	0	0		258	254	−1, 1	$B_{20} = 508\ (506)$
B_{21}^i	−336	−448	0	0	$^7F_1$	391	395	1,−1	
B_{22}^r	382	65	−230	−365		420	425	0, −2	$B_{20} = 468\ (452)$,
B_{22}^i	373	383	0	0					$B_{22}^r = 223\ (38)$
B_{40}	729	845	382	356	$^7F_2$	837	840	2, −1	$B_{20} = 967\ (972)$,
B_{41}^r	686	649	−221	−51		860	865	−2, 2	$B_{40} = 948\ (980)$
B_{41}^i	241	219	−68	−214		911	915	0, 1	

	(b)	(c)	(b')	(c')
B_{42}^r	−244	−187	94	329
B_{42}^i	120	128	−181	−28
B_{43}^r	329	281	−558	−687
B_{43}^i	84	96	−418	−260
B_{44}^r	41	167	396	6
B_{44}^i	100	52	−430	−485
B_{60}	167	175	173	−22
B_{61}^r	161	169	−93	238
B_{61}^i	79	100	268	89
B_{62}^r	230	258	−139	104
B_{62}^i	28	36	154	29
B_{63}^r	−11	−6	127	126
B_{63}^i	54	59	25	−4
B_{64}^r	50	34	38	−31
B_{64}^i	149	−75	95	161
B_{65}^r	−141	−153	19	62
B_{65}^i	−28	−89	−4	108
B_{66}^r	204	−28	−10	195
B_{66}^i	18	−165	157	136
S_2	527	527	527	527
S_4	470	471	470	471
S_6	171	171	171	172

Multiplet	(e)	(f)	(M, M′)	Parameters
	933	937	−1, −2	$B_{20} = 998\ (1012)$,
	959	964	1, −1	$B_{40} = 965\ (988)$
$^7F_3$	1793	1794	3, −3	$B_{60} = 272$,
	1829	1831	−3, 3	$B_{61}^r = 238\ (228)$
	1848	1851	2, 1	
	-	1874	−2, 1	
	1942	1942	1, −2	
	1978	1975	−1, 0	
	2000	1997	0, −1	
$^7F_4$	2811	2811	3, −3	$B_{66}^r = 337\ (–19)$
	2880	2880	2, −3	
	2900	2888	−3, 3	
	2917	2916	0, −1	
	2997	2997	2, −3	
	3049	3047	0, −2	
	3088	3086	1, −1	
	3164	3162	4, 1	
	3183	3181	−4, −3	
$^5D_0$	17238	17239	0	

* The rhombicity ratio: $\kappa = B_{22}^r/B_{20}$ are: 0.20 (b′), 0.344 (c′). The Euler angles are: α = 67.72, β = −103.50, γ = 91.22 (b′); α = 51.95, β = −108.71, γ = 82.84 (c′) (all in degree).

Table S6 YBO$_3$:Eu^{3+}: (a) Allowed crystal-field parameters (CFPs) for the triclinic C_1 point symmetry at Eu^{3+} site; the calculated CFPs values (cm^{-1}) employing: (b) the ECM method and (c) the SPM method along with the rotational invariants, S_k; triclinic ECM/CFPs and SPM/CFPs standardized using the package 3DD - (b′) and (c′); (d) $^{2S+1}L_J$ multiplets of Eu^{3+} ion; (e) experimental[15] and (f) calculated

energy (cm⁻¹) levels; (g) the largest and second largest M_J values of the $|^{2S+1}L_J, M_J\rangle$ levels; and (h) varied values of ECM (SPM)/CFPs are tabulated

| CFPs | | | | | Multiplet | $E_{\exp}$ | E_i^{cal} | M_J | Varied ECM (SPM)/ CFPs |
| | ECM | SPM | 3DD* | | | | | | |
(a)	(b)	(c)	(b′)	(c′)	(d)	(e)	(f)	(g)	(h)
B_{20}	403	478	722	−699	$^7F_0$	0	0	0	0
B_{21}^r	−214	−109	0	0	$^7F_1$	325	324	−1, 1	$B_{20} = 318\ (324),$
B_{21}^i	−142	−136	0	0		352	356	1, 0	$B_{22}^i = -64\ (-92)$
B_{22}^r	334	325	78	−153					
B_{22}^i	−89	−131	0	0		357	363	0, 1	$B_{20} = 318\ (320),$
B_{40}	300	344	−39	−21					$B_{21}^r = -188\ (-152),$
B_{41}^r	−127	−251	481	224					$B_{22}^i = -64\ (-68)$
B_{41}^i	311	317	171	−671					
B_{42}^r	−101	−94	−553	−141	$^7F_2$	803	805	−2, 2	$B_{43}^i = 974\ (942)$
B_{42}^i	440	430	−13	592		825	828	2, −2	
B_{43}^r	146	124	412	55		857	860	0, 1	
B_{43}^i	752	746	270	449		1244	1242	−1, 1	
B_{44}^r	48	29	755	455		1280	1283	1, −1	
B_{44}^i	−655	−628	−5	−317	$^7F_3$	1796	1797	2, −3	$B_{64}^i = -1012\ (358)$
B_{60}	45	103	−382	−364		1859	1863	0, 1	
B_{61}^r	−219	−476	325	−477		1891	1893	−3, 3	
B_{61}^i	−202	−443	−54	−137		1944	1942	1, −1	
B_{62}^r	135	290	7	−402		-	1976	−1,−3	
B_{62}^i	−194	−414	−511	318		2000	2001	0, 1	
B_{63}^r	−55	−111	162	282		2139	2137	−2,−1	$B_{61}^i = -287\ (-381),$
B_{63}^i	−3	10	302	266					$B_{64}^i = -1027\ (364)$
B_{64}^r	−66	−141	−157	−234	$^7F_4$	2466	2474	0, −1	$B_{61}^i = -267\ (-324),$
B_{64}^i	−977	244	322	236					$B_{62}^i = -282\ (-442),$

Parameter	(b)	(b′)	(c)	(c′)
B^r_{65}	−176	−375	221	−393
B^i_{65}	−84	−204	−197	346
B^r_{66}	−211	−477	610	−200
B^i_{66}	33	88	197	−207
S_2	327	328	327	327
S_4	554	555	554	554
S_6	429	428	427	428

(d)	(e)	(f)	(g)	(h)
				$B^i_{64} = -1027\ (264)$
	2776	2771	−3, 3	$B^i_{61} = -226\ (-478)$,
	-	2880	−3, 2	$B^i_{64} = -1008\ (308)$
	2893	2893	−1, 1	
	2995	2995	−2, −3	
	3055	3056	2, −3	
	3099	3095	1, −1	
	-	3098	1, −1	
	3190	3192	0, 1	
$^5D_0$	17198	17200	0, 0	
$^5D_1$	18944	18946	−1, 1	$B^i_{22} = -74\ (-92)$
	18954	18957	1, −1	
	18958	18960	0, 1	
$^5D_2$	21343	21344	−2, 2	
	21354	21357	−1, 2	
	21425	21427	1, −1	
	21456	21457	0, 1	
	21470	21472	2, 0	

* The rhombicity ratio $\kappa = B^r_{22}/B_{20}$ is: 0.11 (b′), 0.22 (c′). The Euler angles are: α = 63.21, β = 33.42, γ = 35.36 (b′); α = 102.01, β = −99.21, γ = 74.33 (c′) (all in degree).

Table S7 $YBO_3:Eu^{3+}$: (a) Allowed crystal-field parameter (CFPs) for the triclinic C_i point symmetry at Eu^{3+} site; the calculated CFPs values (cm^{-1}) employing: (b) the ECM method and (c) the SPM method along with the rotational invariants, S_k; triclinic ECM/CFPs and SPM/CFPs standardized using the package 3DD - (b′) and (c′); (d) $^{2S+1}L_J$ multiplets of Eu^{3+} ion; (e) experimental[15] and (f) calculated energy (cm^{-1}) levels; (g) the largest and second largest M_J values of the $|^{2S+1}L_J, M_J\rangle$ levels; and (h) varied values of ECM (SPM)/CFPs are tabulated

CFPs					Multiplet	E_exp	E_i^cal	M_J	Varied ECM (SPM)/ CFPs
	ECM	SPM	3DD*						
(a)	(b)	(c)	(b′)	(c′)	(d)	(e)	(f)	(g)	(h)
B_{20}	154	157	−674	−669	$^7F_0$	0	0	0	
B_{21}^r	124	254	0	0		320	316	−1, 1	$B_{20} = 90$ (87),
B_{21}^i	−75	−54	0	0	$^7F_1$	352	356	0	$B_{21}^i = -36$ (−24),
B_{22}^r	261	240	−208	−217		408	409	1,−1	$B_{22}^i = -298$ (274)
B_{22}^i	−412	366	0	0	$^5D_0$	17246	17251	0, 6	
B_{40}	−741	−781	499	−266		18949	18950	1, −1	$B_{22}^i = -374$ (323)
B_{41}^r	−185	−153	409	−142	$^5D_1$	18963	18964	0, −1	
B_{41}^i	−368	−354	−604	1071		18974	18977	−1, 1	
B_{42}^r	485	481	18	378					
B_{42}^i	−1433	−1427	1041	−547					
B_{43}^r	−431	−412	233	−1103					
B_{43}^i	−825	−806	10	−706					
B_{44}^r	−119	−77	219	−642					
B_{44}^i	488	547	1417	−273					
B_{60}	−273	−306	63	48					
B_{61}^r	−136	−144	−377	246					
B_{61}^i	240	270	193	333					
B_{62}^r	352	407	−268	−459					
B_{62}^i	156	191	398	612					
B_{63}^r	−443	−505	−223	−325					
B_{63}^i	135	180	382	−159					
B_{64}^r	−38	−41	−267	192					
B_{64}^i	728	−504	522	284					
B_{65}^r	−317	−386	−569	−421					
B_{65}^i	355	388	−88	289					

B_{66}^r	−425	−483	335	392
B_{66}^i	−100	−104	170	95
S_2	329	329	329	329
S_4	924	924	925	924
S_6	468	469	468	469

*The rhombicity ratio $\kappa = B_{22}^r/B_{20}$ is: 0.31 (b′), 0.32 (c′). The Euler angles are: $\alpha = 118.75$, $\beta = -90.52$, $\gamma = -22.04$ (b′); $\alpha = 58.35$, $\beta = -104.88$, $\gamma = 148.31$ (c′) (all in degree).

Table S8 The spherical polar coordinates (R_h, θ_h, ϕ_h) for the O^{2-} ligands around the metal center having site symmetry, C_1 and C_i, in the distorted YO_8 polyhedron in YBO_3 host crystal[41] obtained in the CAS* ($a*//X$, $b//Y$, $c//Z$); see, the text

O^{2-} ion	YBO₃					
	C_1			C_i		
	R_h (Å)	θ_h (°)	ϕ_h (°)	R_h (Å)	θ_h (°)	ϕ_h (°)
O1	2.4667	92.116	−152.660	2.3001	47.4204	−139.8420
O1′	2.4038	70.053	−78.965	2.3001	132.5796	40.1580
O2	2.4448	132.904	−37.346	2.4287	67.5325	82.2336
O2′	2.2762	85.160	26.372	2.4287	112.4675	−97.7664
O3	2.2285	112.706	100.663	2.4332	92.9205	150.1370
O3′	2.4063	48.667	139.640	2.4332	87.0795	−29.8630
O4	2.2929	12.240	−61.156	2.3685	139.3940	−165.5032
O5	2.3329	150.135	−139.140	2.3685	40.6060	14.4968

Table S9 $ZnO:Eu^{3+}$: (a) Allowed crystal-field parameters (CFPs) for the triclinic C_1 point symmetry at the B-site of Eu^{3+}; the calculated CFPs values (cm^{-1}) employing: (b) the ECM method, and (c) the SPM method for the original trigonal C_{3v} symmetry along with the rotational invariants, S_k; (d) adopted CFP values for lower C_1 point symmetry - see the text; (d′) triclinic ECM/CFPs standardized using 3DD package; (e) $^{2S+1}L_J$ multiplets of Eu^{3+} ion; (f) experimental energy (cm^{-1}) levels;[16] calculated energy (cm^{-1}) levels, respectively for: (g) C_{3v} symmetry using the CFPs (see, b and/or c), and (h) C_1 symmetry; and (i) the varied ECM/CFPs are tabulated

CFPs					Multiplet	$E_{\exp}$	E_i^{cal}		Varied ECM/ CFPs
	C_{3v}		C_1				C_{3v}	C_1	
	ECM	SPM	ECM	3DD*					
(a)	(b)	(c)	(d)	(d′)	(e)	(f)	(g)	(h)	(i)
B_{20}	−369	−370	−369	−551	$^7F_0$	0	0	0	
B_{21}^r			214	0	$^7F_1$	318	292	314	$B_{20} = -198$
B_{21}^i			−183	0		351	410	353	$B_{21}^i = -81$
B_{22}^r			111	−109		452	410	449	
B_{22}^i			64	0	$^7F_2$	851	732	849, 906, 966, 1161, 1242	$B_{20} = -210$
B_{40}	1884	1930	1884	419		909	765, 765,		$B_{40} = 1330$
B_{41}^r			563	−212		969	1433, 1433		$B_{43}^r = 1496$
B_{41}^i			178	938		1162			
B_{42}^r			−244	374		1238			
B_{42}^i			67	1831	$^7F_3$	1882	1805, 1805, 1885, 1994, 2016, 2016, 2192	1843, 1877, 1935, 1963, 2006, 2063, 2240	$B_{60} = 987$
B_{43}^r	2025	2004	2025	−201		1931			$B_{62}^r = 323$
B_{43}^i			24	−554		1961			
B_{44}^r			251	1026		2003			
B_{44}^i			−52	−672	$^7F_4$	2844	2351, 2853, 2862, 2862, 3136, 3136, 3207, 3207, 3224	2419, 2841, 2851, 2992, 3120, 3177, 3206, 3211, 3296	
B_{60}	1128	1091	1128	−917		2994			
B_{61}^r			157	578		3118			
B_{61}^i			13	56	$^5D_0$	17135	17132	17133	
B_{62}^r			220	−30	$^5D_1$	18880	18879, 18912, 18912	18880, 18912, 18911	$B_{21}^r = 175$
B_{62}^i			−2	285					$B_{22}^r = 152$
B_{63}^r	−765	−810	−765	−410	$^5D_2$	21292	21326, 21326, 21414, 21417, 21417,	21293, 21304, 21379, 21391, 21407	$B_{41}^r = 104$
B_{63}^i			17	−59		21377			
B_{64}^r			7	−526		21406			
B_{64}^i			−78	468	$^5D_3$	24195	24233, 24246, 24259, 24259, 24303, 24309,	24185, 24193, 24209, 24211, 24251, 24255,	$B_{61}^r = 120$
B_{65}^r			−134	175					$B_{62}^r = 182$

B_{65}^i			10	−362			24309	24257
B_{66}^r	875	859	875	−564				
B_{66}^i			−20	−315	${}^5L_6$	24616	24693, 24696, 24696, 24711, 24711, 24720, 25350, 25350, 25383, 25422, 25422, 25451, 25545	24607, 24612, 24618, 24622, 24626, 24634, 25028, 25165, 25266, 25300, 25310, 25330, 25422
S_2	165	165	256	256		25027		
S_4	1143	1143	1188	1188		25162		
S_6	553	553	567	566				

*The rhombicity ratio $\kappa = B_{22}^r/B_{20}$ is: 0.197 (d′). The Euler angles are: $\alpha = 51.01$, $\beta = 28.26$, $\gamma = 42.02$ (d′) (all in degree).

Table S10 The spherical polar coordinates (R_h, θ_h, ϕ_h) for the O^{2-} ligands around Zn^{2+} center in the distorted ZnO_4 tetrahedron having C_{3v} symmetry in ZnO

O^{2-} ion	ZnO		
	R_h (Å)	θ_h (°)	ϕ_h (°)
O1	2.001	0	0
O2	1.9735	107.8325	120
O3	1.9735	107.8325	240
O4	1.9735	107.8325	0

Table S11 ZnO:Eu^{3+}: (a) Allowed crystal-field parameters (CFPs) for the triclinic C_1 point symmetry at the A-site of Eu^{3+}; (b) the adopted CFPs values (cm^{-1}) for C_1 symmetry -see, the text; (c) ${}^{2S+1}L_J$ multiplets of Eu^{3+} ion; energy levels: (d) experimental,[16] and (e) calculated (cm^{-1}); (f) the varied CFPs are tabulated

CFPs C_1		Multiplets	E_{exp}	E_i^{cal} C_1	Varied CFPs
(a)	(b)	(c)	(d)	(e)	(f)
B_{20}	−369	${}^7F_0$	0	0	
B_{21}^r	214	${}^7F_1$	299	298	$B_{20} = -222$, $B_{21}^i = -95$
B_{21}^i	−183		369	371	
			446	443	

B_{22}^{r}	111	$^7F_2$	993	809, 863, 991, 1085, 1176	$B_{20} = -265$, $B_{40} = 1470$,
B_{22}^{i}	64				$B_{43}^{r} = 1966$
B_{40}	1884	$^7F_3$	1887	1783, 1815, 1890, 1950, 1997, 2055, 2126	$B_{60} = 1016$, $B_{62}^{r} = 304$
B_{41}^{r}	563				
B_{41}^{i}	178	$^7F_4$	2713 2921	2312, 2714, 2797, 2919, 3032, 3093, 3152, 3180, 3211	
B_{42}^{r}	−244	$^5D_0$	17271	17274	
B_{42}^{i}	67	$^5D_1$	19001	18996, 19003, 19020	$B_{21}^{r} = 188$, $B_{22}^{r} = 144$
B_{43}^{r}	2025	$^5D_2$	21515	21482, 21517, 21545, 21565, 21576	$B_{41}^{r} = 115$
B_{43}^{i}	24	$^5L_6$	25368 [25380][a]	25000, 25008, 25022, 25047, 25061, 25090, 25371, 25402, 25692, 25708, 25717, 25756, 25831	$B_{61}^{r} = 129$, $B_{62}^{r} = 194$
B_{44}^{r}	251				
B_{44}^{i}	−52				
B_{60}	1128				
B_{61}^{r}	157				
B_{61}^{i}	13				
B_{62}^{r}	220				
B_{62}^{i}	−2				
B_{63}^{r}	−765				
B_{63}^{i}	17				
B_{64}^{r}	7				
B_{64}^{i}	−78				
B_{65}^{r}	−134				
B_{65}^{i}	10				
B_{66}^{r}	875				
B_{66}^{i}	−20				

a) $^5L_6 \rightarrow {}^7F_0$ transition energy in ZnO:Eu^{3+} is 3.147 eV = 25380 cm^{-1} [16]

Table S12 YAl$_3$(BO$_3$)$_4$:Eu^{3+}: (a) Allowed crystal-field parameters (CFPs) for the trigonal D$_3$ point symmetry at Eu^{3+} site; the calculated CFPs values obtained using: (b) the LSC method[11] and (c) ADS method;[11] and (d) the ECM and (e) SPM method along with the rotational invariants, S_k; (f) $^{2S+1}L_J$ multiplets of Eu^{3+} ion; (g) experimental energy (cm^{-1}) levels;[11] (h) calculated energy (cm^{-1}) levels using the ECM/CFPs; and (i) the largest and second largest M_J values of the $|^{2S+1}L_J, M_J\rangle$ levels are tabulated

| CFPs | | | | Multiplet | E_{exp} | E_i^{cal} | M_J |
| | LSC | ADS | ECM | SPM | | | | |
(a)	(b)	(c)	(d)	(e)	(f)	(g)	(h)	(i)
B_{20}	467	493	407	408	$^7F_0$	0 (A$_1$)	0	0
B_{40}	−1007	−1809	−1372	−1338	$^7F_1$	323 (E)	325 (E)	(±1, ∓1)
B_{43}^r	−936	−677	−943	−965		453 (A$_2$)	450	0
B_{60}	516	−488	565	562		931 (E)	930 (E)	(∓1, ∓2)
B_{63}^r	1902	788	57	54	$^7F_2$	1049 (E)	1049 (E)	(±2, ∓1)
B_{66}^r	520	860	−230	−240		---	1204	(0, 3)
						1821 (A$_2$)	1821	(−3, 3)
S_2	208	220	182	182		---	1901	(3, −3)
S_4	554	682	638	637	$^7F_3$	1939 (E)	1938 (E)	(±1, ∓1)
S_6	786	477	182	183		1965 (E)	1964 (E)	(∓2, ±1)
						2063 (A$_2$)	2058	(0,−3)
						2536 (E)	2540	(0, −3)
						2596 (E)	2606 (E)	(±4, ±1)
					$^7F_4$	---	2766 (E)	(∓1, ±1)
						2907 (A$_2$)	2908	(3, −3)
						--	3003	(−3, 3)
						3009 (E)	3011 (E)	(∓2, ±2)

						$^7F_5$		3804 (E)	$(\pm5, \mp5)$
								3820	$(0, -3)$
								3823 (E)	$(\pm1, \mp1)$
								3889 (E)	$(\mp2, \mp5)$
								3983	$(-3, 3)$
							3997 (A_2)	4000	$(3, -3)$
							4092 (E)	4089 (E)	$(\mp4, \mp1)$
						$^7F_6$	4863 (E)	4863 (E)	$(\mp4, \mp1)$
							---	4875	$(3, -3)$
							4893 (E)	4895 (E)	$(\mp2, \mp5)$
							4905 (A_2)	4908	$(-3, 3)$
							----	5086	$(0, 3)$
							5087 (E)	5088 (E)	$(\mp1, \mp4)$
							5166 (E)	5169 (E)	$(\pm5, \pm2)$
							5353 (A_2)	5351	$(6, -6)$
							---	5352	$(-6, 6)$
						$^5D_0$	17215 (A_1)	17219	0
						$^5D_1$	18965 (E)	18966 (E)	$(\pm1, \mp1)$
							19001 (A_2)	19002	$(0, 6)$
						$^5D_2$	21415 (A_1)	21414	$(0, -3)$
							21418 (E)	21420 (E)	$(\pm2, \mp1)$
							21489 (E)	21490 (E)	$(\mp1, \pm2)$
						$^5D_3$	24235 (E)	24237 (E)	$(\mp1, \pm2)$
							24292 (E)	24298 (E)	$(\mp2, \pm1)$
							24344 (A_2)	24347	$(0, -3)$
							24405 (A_1)	24402	$(-3, 3)$
							24618 (A_2)	24614	$(0, 3)$

							24877 (A$_2$)	24877	(6, −6)
							---	24893	(−6, 6)
							24934 (E)	24937	(0, 6)
						$^5L_6$	25026 (E)	24939 (E)	($\mp$1, ±2)
							25117 (E)	25013 (E)	(±3, ±5)
							25215 (A$_2$)	25225	(3, −3)
							----	25262	(−3, 3)
							----	25338 (E)	($\mp$4, $\mp$1)
							25341 (E)	25346 (E)	($\mp$5, $\mp$2)

Table S13 The spherical polar coordinates (R_h, θ_h, ϕ_h) for the O^{2-} ligands around metal center in the distorted trigonal (D_3) YO_6 polyhedron in $YAl_3(BO_3)_4$; Φ (= 8°) is the distortion angle. For explanations, see, text

O^{2-} ion	$YAl_3(BO_3)_4$		
	R_h (Å)	θ_h (°)	ϕ_h (°)
O1	2.154	60.76	90 + Φ
O1′	2.154	119.24	90 − Φ
O2	2.154	60.76	210 + Φ
O2′	2.154	119.24	210 − Φ
O3	2.154	60.76	330 + Φ
O3′	2.154	119.24	330 − Φ

References: (see main text)