Supplementary materials for article:

"Compound-tunable embedding potential method: Analysis of pseudopotentials for Yb in YbF_2, YbF_3, $YbCl_2$ and $YbCl_3$ crystals"

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Compound-tunable embedding potential (CTEP) method developed in [Lomachuk *et al., PCCP*, 2020, **22**, 17922; Maltsev *et al., PRB*, 2021, **103**, 205105] to describe electronic structure of fragments and point defects in materials is applied to crystals containing periodically arranged lanthanide atoms, which can have open $4f$ shell. We consider YbF_2, YbF_3, $YbCl_2$, and $YbCl_3$ crystals for the pilot CTEP studies such that $4f$ electrons are not treated explicitly at the CTEP generation stages. Instead, the pseudopotentials with 60 and 59 electrons in core for Yb(II) and Yb(III), correspondingly, are applied and the latter treats the "$4f$-hole in core". At the final stage, the two-component embedded cluster study of fragments of $YbHal_2$ and $YbHal_3$ crystals (Hal = F, Cl) is performed using the CTEP method and relativistic pseudopotential with 28 electrons in core for central Yb atom. Remarkable agreement of the electronic densities within the $YbHal_2$ fragments with those of the original periodic crystal calculation is demonstrated. The calculated internuclear distances between the central Yb and nearest halide atoms are in pretty good agreement with the experimental data, the deviations are within 0.015 Å for all the studied crystals. Thus, the overall accuracy for the crystal characteristics evaluated with using CTEP in the combined periodic-structure and embedded cluster study is comparable with that of Yb-containing molecular calculations.

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Table 1 The basic sets for ytterbium atom that were used in this work. The Gaussians that were used only for cluster calculations are marked in parentheses.

Basis sets for ytterbium, which were made for following PPs										
	lc-PP				mc-PP				sc-PP	
	2ve-PP		3ve-PP		10ve-PP		11ve-PP		42ve-PP	
	Exp	Coeff	Exp	Coeff	Exp	Coeff	Exp	Coeff	Exp	Coeff
s	1.703353	0.032943	1.606205	0.032835	5.66229	-0.2395634	23.69822	-0.86659	23.69822	-0.86659
	0.359455	-0.477703	0.735687	0.011886	3.05589	0.8934967	18.30547	1.59105	18.30547	1.59105
					1.07726	-0.6980144				
							6.02665	1.0000000	6.02665	1.0000000
							2.93645	1.0000000	2.93645	1.0000000
							1.38490	1.0000000	1.38490	1.0000000
					0.44751	1.0000000	0.85837	1.0000000	0.85837	1.0000000
	0.298340	1.0000000	0.39304	1.0000000	0.29834	1.0000000	0.39304	1.0000000	(0.39304)	(1.0000000)
	0.132600	1.0000000			0.13260	1.0000000			(0.11464)	(1.0000000)
p	0.292208	-0.201665	0.653783	0.005863	4.07728	0.0914961	23.92973	-0.034234	23.92973	-0.034234
	0.146922	0.097861	0.336487	-0.253364	3.39773	-0.2494782	13.21051	-0.42757	13.21051	-0.42757
					0.78282	0.4112846	10.39351	1.09428	10.39351	1.09428
							12.51956	1.0000000	12.51956	1.0000000
							4.02239	1.0000000	4.02239	1.0000000
					1.64606	1.0000000	1.72446	1.0000000	1.72446	1.0000000
					0.34423	1.0000000	0.70647	1.0000000	0.70647	1.0000000
	0.13929	1.0000000	0.29693	1.0000000	0.13929	1.0000000	0.29693	1.0000000	(0.29693)	(1.0000000)
d	2.392979	-0.013914	3.20980	-0.042259			54.12548	-0.46786E-02	54.12548	-0.46786E-02
	0.825371	0.276282	1.371021	0.137661			9.089210	0.19280	9.089210	0.19280
	0.284682	0.541616	0.585579	0.393576			4.42200	0.21885	4.42200	0.21885
							2.14170	1.0000000	2.14170	1.0000000
					0.61623	1.0000000	0.77727	1.0000000	0.77727	1.0000000
	0.18155	1.0000000	0.32550	1.0000000	0.18155	1.0000000	0.32550	1.0000000	(0.32550)	(1.0000000)
f							132.7621	0.59655E-02	132.7621	0.59655E-02
							43.76773	0.03721	43.76773	0.03721
							16.92991	0.11234	16.92991	0.11234
							6.97518	0.17748	6.97518	0.17748
							2.80817	1.0000000	2.80817	1.0000000
					1.02851	1.0000000	1.02851	1.0000000	1.02851	1.0000000
	0.31000	1.0000000	0.31000	1.0000000	0.31000	1.0000000	0.310000	1.0000000	(0.310000)	(1.0000000)

Table 2 Large-core pseudopotentials for ytterbium atom that were used in this work. For compound-tunnable lc-PP, exponent with (*) equals 0.2267468 and 0.2484787 for YbF_2 and $YbCl_2$ compounds, accordingly; exponent with (**) equals 0.4485319 and 0.4492845 for YbF_3 and $YbCl_3$ compounds, accordingly.

lc-PP for Yb						
	2ve-PP			3ve-PP		
	n_{core} = 68	l_{max} = 4		n_{core} = 67	l_{max} = 4	
	Exponent	r^n	Coefficient	Exponent	r^n	Coefficient
g	12.1426975	1	-15.2386012	1.1833077	1	-12.4579302
	3.6608059	1	-14.5775174			
	0.9869034	1	-7.9349017			
	0.3559306	1	-0.8397914			
s-g	0.1198414	0	10.0000000	0.4752020	0	10.0000000
	8.2657933	1	22.7174604	1.1833077	1	12.4579302
	1.4990175	1	11.7793783			
	0.1876474	2	-2.0249546			
p-g	0.2757268	0	9.0000000	0.2839004 (**)	0	9.0000000
	9.9771245	1	19.3438417	1.1833077	1	12.4579302
	2.3035432	1	13.5516361			
	0.5285219	1	3.1375203			
d-g	2.5235783	0	7.0000000	5.9601207	0	7.0000000
	9.9798004	1	14.8276980	1.1833077	1	12.4579302
	2.2132429	1	15.4139348			
	0.1344036	1	-0.2753461			
f-g	0.6068395 (*)	0	4.0000000	1.0482995	0	4.0000000
	12.1426975	1	15.2386012	1.1833077	1	12.4579302
	3.6608059	1	14.5775174			
	0.9869034	1	7.9349017			
	0.3559306	1	0.8397914			

Table 3 Medium-core pseudopotentials for ytterbium atom that were used in this work.

mc-PP for Yb						
	n_{core} = 60	10ve-PP l_{max} = 4		n_{core} = 59	11ve-PP l_{max} = 4	
	Exponent	r^n	Coefficient	Exponent	r^n	Coefficient
g	28.0911153	1	-20.9275297	28.5078345	1	-17.3720028
	20.5917581	1	73.3189526	20.6456816	1	62.6511064
	15.1315885	1	-56.3156303	14.7718856	1	-49.4713307
	3.7895130	1	-12.2315386	3.8867871	1	-11.7762364
	1.3010428	1	-4.1563613	1.3774110	1	-3.0004553
	0.8542142	1	0.7630462	0.4491757	1	-0.3443483
	0.5583110	1	-0.8963727	0.2814416	1	0.0775801
	0.1848613	1	-0.0295597	0.1695274	1	-0.0186098
	0.1158813	2	0.0001565	0.0981397	2	0.0003351
	0.0549310	2	-0.0000092	0.0409101	2	-0.0000147
	0.0174546	2	-0.0000023	0.0132964	2	-0.0000008
s-g	14.8864634	0	10.0000000	15.0652965	0	10.0000000
	20.3783278	1	15.1304038	21.2163730	1	14.9214061
	20.3160267	2	-216.7470797	20.6023494	2	-215.7768483
	14.8864650	2	299.7971581	15.0652965	2	301.0973948
	3.5889931	2	163.6607952	3.6049965	2	164.7967825
	2.6702312	2	-92.6457163	2.6870671	2	-92.2349851
	1.9593045	2	20.6998650	1.9747408	2	18.6983031
	0.3589311	2	0.0699011	0.3170567	2	0.0104376
	0.2368430	2	-0.0187870	0.2056531	2	-0.0035788
p-g	2.3975801	0	9.0000000	19.6090345	0	9.0000000
	29.5204845	1	32.1195478	19.4297994	1	-32.4109446
	20.6721605	1	-91.1075481	9.6150372	1	96.3648985
	14.3684138	1	75.0601845	1.1176714	1	-47.9409481
	3.2254444	2	56.3095791	3.7688912	2	131.1092861
	2.3975801	2	-22.3039990	2.1069201	2	8.5017036
	1.7503254	2	3.3613521	1.5164804	2	36.9668225
	0.5224168	2	0.1343950	1.0708112	2	14.4977366
	0.0617217	2	-0.0004131	0.7559869	2	-0.0897142
	0.0371704	2	0.0002820	0.5237791	2	0.0037375
	0.0222136	2	-0.0000693	0.0375115	2	0.0000177
d-g	38.8596928	0	7.0000000	38.7587374	0	7.0000000
	10.5899224	1	91.1323893	10.7082326	1	90.1660572
	4.7884406	1	-16.2551241	4.6255379	1	-1.5100076
	3.1823270	1	279.0564279	3.1697505	1	284.6892687
	1.4882586	1	-338.4892772	1.6729490	1	-358.0786118
	1.7493084	2	374.9429646	1.8922597	2	377.0266229
	0.8737250	2	5.8544893	0.7763756	2	1.2774933
	0.5843255	2	-0.8087249	0.5092806	2	-0.2560451
	0.0749260	2	-0.0030742	0.0842930	2	-0.0021938
f-g	5.3972773	0	4.0000000	4.6451229	0	4.0000000
	1.8280874	1	26.7002567	1.4621505	1	24.6533742
	0.6501774	1	1.4901331	0.5664228	1	0.4741747
	0.2052912	1	0.1432826	0.0443210	1	0.0047029
	0.0188892	1	-0.0000447	0.0320904	1	-0.0019114

Table 4 Small-core pseudopotentials for ytterbium atom that were used in this work. Spin-orbit part was not included in periodic calculations, but it was used in clusters calculations.

lc-PP for Yb							
sc-PP, 42ve-PP							
	$n_{core} = 28$	$l_{max} = 3$			Spin-orbit $l^{so}_{max} = 4$		
	Exponent	r^n	Coefficient		Exponent	r^n	Coefficient
f	393.3778330	1	-13.9350552	p-SpinOrbit	36.3960764	1	-2.8983573
	290.9108996	1	42.9982597		26.0036764	1	25.7649421
	217.2246880	1	-34.5780535		18.3131485	1	-49.7867563
	47.3835015	1	-9.9869518		11.1848920	1	38.5415042
	16.5563358	1	-1.7523321		6.0116579	1	-14.2979100
	2.6083357	2	0.0634998		3.1974227	1	5.5571864
	0.9133649	2	0.0061097		1.6111050	1	-2.0956344
	0.3587417	2	-0.0000728		0.7862135	1	0.8606535
					0.3667289	1	-0.4018655
s-f	134.0286034	0	6.0000000		0.1802831	1	0.2781659
	37.4815435	1	431.1583378		0.0807875	1	-0.1532202
	26.6166901	1	-366.7463438		0.0659333	2	0.0285462
	26.8473429	2	-1089.1177110		0.0406617	2	-0.0073401
	19.2682415	2	3653.6259232		0.0250163	2	0.0010489
	13.8715524	2	-2931.4492940				
	9.8967735	2	1621.0237256	d-SpinOrbit	71.1494532	1	0.2475452
	6.9897690	2	-683.6851818		20.1908161	1	-1.6558115
	4.8791455	2	220.3474487		14.8929508	1	1.5851440
	3.3520194	2	-51.7868352		9.3696110	2	0.2289516
	2.2480714	2	7.7052612		2.8854225	2	-0.0064624
	1.4062180	2	-0.5132751		1.2522332	2	-0.0020983
					0.0234481	2	0.0000004
p-f	23.4359935	0	5.0000000				
	36.3960764	1	166.0036181	f-SpinOrbit	393.3778330	1	1.8156013
	26.0036764	1	-422.8378703		290.9108996	1	-5.9812102
	18.3131485	1	383.8257841		217.2246880	1	5.3013149
	11.1848920	1	-97.9398029		47.3835015	1	0.0645827
	6.0116579	1	23.1230723		16.5563358	1	0.0192377
	3.1974227	1	-7.3316100		2.6083357	2	0.0029396
	1.6111050	1	2.3456439		0.9133649	2	0.0006479
	0.7862135	1	-0.8965902		0.3587417	2	0.0000196
	0.3667289	1	0.3960258				
	0.1802831	1	-0.2663962				
	0.0807875	1	0.1452787				
	0.0659333	2	-0.0270102				
	0.0406617	2	0.0069557				
	0.0250163	2	-0.0009977				
d-f	1.9152345	0	3.0000000				
	71.1494532	1	16.3202169				
	20.1908161	1	-43.4625717				
	14.8929508	1	62.8809242				
	9.3696110	2	-53.2260833				
	2.8854225	2	-7.7122479				
	1.2522332	2	-0.1031502				
	0.0234481	2	-0.0000008				