

**Supplementary Information for:**  
**Neural network-based pseudopotential: development of a  
transferable local pseudopotential**

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# 1 The parameters for NNLP optimization

For the sake of simplicity, we used the same set of many adjustable parameters for all elements.  $w_1$ ,  $w_3$ , and  $w_4$  were set to 100, 10, and 10 for all elements, respectively. Only  $r_c$ ,  $r_t$ ,  $d_{\max}$ , and  $w_2$  were set differently for each element.  $r_c$  was set as the radius at the outermost maximum of the valence wavefunctions of all-electron calculations, and the train radius  $r_t$  was chosen to be 0.5 bohr larger than  $r_c$ .  $w_2$  was optimized differently for each element to decrease the sum of errors in eigenvalues using the Bayesian optimization method.  $d_{\max}$  was used as 10 or 30 for all elements. The detailed values for  $r_c$ ,  $d_{\max}$ , and  $w_2$  are shown in Table S1.

Table S 1: Input parameters to generate the NNLP of each element.

Element	$r_c$ (Å)	$d_{\max}$	$w_2$
Li	3.1311	10	6.1640
Be	2.0497	10	19.4152
B	1.5629	10	19.3460
C	1.1900	10	1.0000
Na	3.2482	10	3.0989
Mg	2.5171	10	13.3235
Al	2.6407	10	18.5779
Si	2.1403	10	35.0619
P	1.8293	30	36.8689
S	1.5959	30	36.8677
Cl	1.4202	10	10.3095
Ar	1.2887	10	1.0052
K	4.0211	10	3.8104
Ca	3.3343	10	5.0384
Ga	2.5244	10	6.1550
Ge	2.2040	10	6.2662
As	1.9729	10	13.3238
Se	1.7961	30	3.5700
Br	1.6630	30	18.0291
Kr	1.5411	10	12.2823

## 2 Effect of the unbound d-state on the accuracy in bulk property calculations

For elements with unbound d-states, the p-d coupling may be overestimated in bulk structures if the d-state is not properly considered. So, the reliable NCPPs of these elements should be generated such that the d-state becomes an unbound state. We generated NCPPs for Ga and Ge by considering only the s- and p-states like in the case of NNLP in this work. Here,  $r_c$  was set to the same value with that of NNLP for each element. And bulk properties were calculated by KS-DFT with these NCPPs (NCPP\* in Table S2), and compared the results with those of NNLP.

Table S 2: Bulk properties calculated by KS-DFT with NCPP, NCPP\*, and NNLP. LDA was used as the exchange-correlation functional.

System	PP	$V_0$ ( $\text{\AA}^3$ )	$B_0$ (GPa)	$B'_0$	$E_c$ (eV)
Ga orthorhombic	NCPP	20.11	53.1	5.21	-3.178
	NCPP*	15.91	86.8	4.78	-3.993
	NNLP	15.22	90.3	4.92	-4.057
Ge diamond	NCPP	23.20	65.0	4.82	-4.489
	NCPP*	20.78	81.9	4.69	-5.001
	NNLP	20.88	77.9	4.46	-4.942

### 3 Electronic configuration test

To evaluate the transferability of NNLPs to other electronic configuration test, we performed atomic calculations for the following three electronic configurations: the ground, excited, and cationic states. The excited states were selected as the state in which one electron has transitioned from the s-state to the p-state. For Li and Na,  $2s^{0.5}2p^0$  and  $3s^{0.5}3p^0$  configurations were used as the cationic states, respectively, because they have only one valence electron in the ground state. For all evaluated elements, spin-unpolarized calculations were performed.

Table S 3: The eigenvalues and excitation energies for the three electronic configurations calculated by all-electron potential, NNLP, OEPP, and BLPS.

Element	Configuration		AE (eV)	NNLP (eV)	OEPP (eV)	BLPS (eV)
Li	$2s^12p^0$	$\varepsilon_0$	-2.877	-2.878	-3.416	-2.976
		$\varepsilon_1$	-1.128	-0.738	-0.364	-0.740
		$\Delta E$	0.000	0.000	0.000	0.000
	$2s^02p^1$	$\varepsilon_0$	-3.344	-3.657	-3.344	-3.802
		$\varepsilon_1$	-1.583	-1.492	-1.519	-1.538
		$\Delta E$	1.754	2.173	3.398	2.283
	$2s^{0.5}2p^0$	$\varepsilon_0$	-5.322	-5.288	-6.549	-5.445
		$\varepsilon_1$	-3.364	-2.662	-2.387	-2.660
		$\Delta E$	2.045	2.039	2.476	2.102
Be	$2s^22p^0$	$\varepsilon_0$	-5.606	-5.598	-6.018	N/A
		$\varepsilon_1$	-2.105	-0.893	-0.632	N/A
		$\Delta E$	0.000	0.000	0.000	N/A
	$2s^12p^1$	$\varepsilon_0$	-6.141	-7.284	-8.321	N/A
		$\varepsilon_1$	-2.604	-2.018	-1.927	N/A
		$\Delta E$	3.519	4.984	5.909	N/A
	$2s^12p^0$	$\varepsilon_0$	-13.647	-13.578	-14.795	N/A
		$\varepsilon_1$	-9.824	-7.193	-7.004	N/A
		$\Delta E$	9.520	9.490	10.281	N/A
Na	$3s^13p^0$	$\varepsilon_0$	-2.823	-2.810	-2.814	N/A
		$\varepsilon_1$	-0.774	-0.777	-0.786	N/A
		$\Delta E$	0.000	0.000	0.000	N/A
	$3s^03p^1$	$\varepsilon_0$	-3.698	-3.733	-3.728	N/A
		$\varepsilon_1$	-1.495	-1.503	-1.506	N/A
		$\Delta E$	2.120	2.124	2.117	N/A
	$3s^{0.5}3p^0$	$\varepsilon_0$	-5.217	-5.224	-5.224	N/A
		$\varepsilon_1$	-2.676	-2.674	-2.686	N/A
		$\Delta E$	2.003	2.000	2.002	N/A
Mg	$3s^23p^0$	$\varepsilon_0$	-4.787	-4.805	-4.821	-4.806
		$\varepsilon_1$	-1.377	-1.372	-1.267	-1.338
		$\Delta E$	0.000	0.000	0.000	0.000
	$3s^13p^1$	$\varepsilon_0$	-5.770	-5.784	-5.920	-5.832
		$\varepsilon_1$	-2.129	-2.135	-2.087	-2.123
		$\Delta E$	3.523	3.540	3.692	3.588
	$3s^13p^0$	$\varepsilon_0$	-11.527	-11.467	-11.597	-11.498
		$\varepsilon_1$	-7.164	-7.085	-6.953	-7.023
		$\Delta E$	8.074	8.068	8.134	8.080

Al	3s <sup>2</sup> 3p <sup>1</sup>	$\varepsilon_0$	-7.836	-8.056	-8.084	-8.197
		$\varepsilon_1$	-2.789	-2.785	-2.639	-2.707
		$\Delta E$	0.000	0.000	0.000	0.000
	3s <sup>1</sup> 3p <sup>2</sup>	$\varepsilon_0$	-8.669	-8.955	-9.085	-9.182
		$\varepsilon_1$	-3.411	-3.444	-3.337	-3.402
		$\Delta E$	5.155	5.396	5.602	5.640
	3s <sup>1</sup> 3p <sup>1</sup>	$\varepsilon_0$	-15.724	-15.931	-16.022	-16.192
		$\varepsilon_1$	-9.849	-9.757	-9.556	-9.700
		$\Delta E$	11.666	11.892	11.942	12.085
Si	3s <sup>2</sup> 3p <sup>2</sup>	$\varepsilon_0$	-10.884	-11.505	-11.838	-11.892
		$\varepsilon_1$	-4.169	-4.167	-3.881	-4.081
		$\Delta E$	0.000	0.000	0.000	0.000
	3s <sup>1</sup> 3p <sup>3</sup>	$\varepsilon_0$	-11.639	-12.390	-12.924	-12.892
		$\varepsilon_1$	-4.746	-4.810	-4.596	-4.774
		$\Delta E$	6.807	7.462	8.148	7.969
	3s <sup>2</sup> 3p <sup>1</sup>	$\varepsilon_0$	-19.109	-19.647	-19.871	-20.073
		$\varepsilon_1$	-11.753	-11.581	-11.073	-11.464
		$\Delta E$	7.829	7.758	7.354	7.653
P	3s <sup>2</sup> 3p <sup>3</sup>	$\varepsilon_0$	-14.021	-15.142	-16.159	-15.649
		$\varepsilon_1$	-5.603	-5.598	-5.177	-5.483
		$\Delta E$	0.000	0.000	0.000	0.000
	3s <sup>1</sup> 3p <sup>4</sup>	$\varepsilon_0$	-14.728	-16.022	-17.366	-16.676
		$\varepsilon_1$	-6.157	-6.243	-5.933	-6.184
		$\Delta E$	8.496	9.665	11.212	10.332
	3s <sup>2</sup> 3p <sup>2</sup>	$\varepsilon_0$	-23.325	-24.378	-25.275	-24.834
		$\varepsilon_1$	-14.306	-14.119	-13.415	-13.874
		$\Delta E$	9.821	9.738	9.169	9.559
S	3s <sup>2</sup> 3p <sup>4</sup>	$\varepsilon_0$	-17.284	-19.170	-21.006	N/A
		$\varepsilon_1$	-7.112	-6.836	-6.561	N/A
		$\Delta E$	0.000	0.000	0.000	N/A
	3s <sup>1</sup> 3p <sup>5</sup>	$\varepsilon_0$	-17.959	-20.171	-22.327	N/A
		$\varepsilon_1$	-7.653	-7.529	-7.361	N/A
		$\Delta E$	10.240	12.490	14.710	N/A
	3s <sup>2</sup> 3p <sup>3</sup>	$\varepsilon_0$	-27.633	-29.260	-31.199	N/A
		$\varepsilon_1$	-16.884	-16.152	-15.834	N/A
		$\Delta E$	11.863	11.372	11.067	N/A
Ca	4s <sup>2</sup> 4p <sup>0</sup>	$\varepsilon_0$	-3.869	-3.759	-3.781	N/A
		$\varepsilon_1$	-1.441	-1.601	-1.662	N/A
		$\Delta E$	0.000	0.000	0.000	N/A
	4s <sup>1</sup> 4p <sup>1</sup>	$\varepsilon_0$	-4.548	-4.359	-4.333	N/A
		$\varepsilon_1$	-1.989	-2.073	-2.109	N/A
		$\Delta E$	2.491	2.219	2.170	N/A
	4s <sup>1</sup> 4p <sup>0</sup>	$\varepsilon_0$	-9.175	-9.031	-9.023	N/A
		$\varepsilon_1$	-6.066	-6.249	-6.341	N/A
		$\Delta E$	6.463	6.327	6.339	N/A

Ga	4s <sup>2</sup> 4p <sup>1</sup>	$\varepsilon_0$	-9.174	-9.155	-9.158	-8.911
		$\varepsilon_1$	-2.738	-2.744	-2.698	-2.671
		$\Delta E$	0.000	0.000	0.000	0.000
	4s <sup>1</sup> 4p <sup>2</sup>	$\varepsilon_0$	-10.280	-10.394	-10.331	-10.010
		$\varepsilon_1$	-3.500	-3.532	-3.490	-3.435
		$\Delta E$	6.613	6.639	6.658	6.414
	4s <sup>1</sup> 4p <sup>1</sup>	$\varepsilon_0$	-17.753	-17.988	-17.795	-17.416
		$\varepsilon_1$	-10.200	-10.293	-10.136	-10.063
		$\Delta E$	13.338	13.427	13.354	13.042
As	4s <sup>2</sup> 4p <sup>3</sup>	$\varepsilon_0$	-14.703	-14.681	-14.886	-14.721
		$\varepsilon_1$	-5.347	-5.343	-5.298	-5.311
		$\Delta E$	0.000	0.000	0.000	0.000
	4s <sup>1</sup> 4p <sup>4</sup>	$\varepsilon_0$	-15.587	-15.700	-15.879	-15.669
		$\varepsilon_1$	-5.980	-6.004	-5.972	-5.969
		$\Delta E$	9.483	9.517	9.751	9.558
	4s <sup>2</sup> 4p <sup>2</sup>	$\varepsilon_0$	-23.607	-23.555	-23.776	-23.627
		$\varepsilon_1$	-13.545	-13.473	-13.424	-13.476
		$\Delta E$	9.325	9.292	9.245	9.277
Se	4s <sup>2</sup> 4p <sup>4</sup>	$\varepsilon_0$	-17.485	-17.480	-17.574	N/A
		$\varepsilon_1$	-6.661	-6.688	-6.674	N/A
		$\Delta E$	0.000	0.000	0.000	N/A
	4s <sup>1</sup> 4p <sup>5</sup>	$\varepsilon_0$	-18.318	-18.421	-18.469	N/A
		$\varepsilon_1$	-7.268	-7.319	-7.306	N/A
		$\Delta E$	10.939	10.947	11.033	N/A
	4s <sup>2</sup> 4p <sup>3</sup>	$\varepsilon_0$	-27.081	-27.065	-27.181	N/A
		$\varepsilon_1$	-15.589	-15.575	-15.582	N/A
		$\Delta E$	11.009	11.021	11.017	N/A
Br	4s <sup>2</sup> 4p <sup>5</sup>	$\varepsilon_0$	-20.322	-20.487	-20.725	N/A
		$\varepsilon_1$	-8.007	-8.020	-7.979	N/A
		$\Delta E$	0.000	0.000	0.000	N/A
	4s <sup>1</sup> 4p <sup>6</sup>	$\varepsilon_0$	-21.118	-21.418	-21.637	N/A
		$\varepsilon_1$	-8.598	-8.646	-8.615	N/A
		$\Delta E$	12.418	12.620	12.886	N/A
	4s <sup>2</sup> 4p <sup>4</sup>	$\varepsilon_0$	-30.592	-30.736	-31.001	N/A
		$\varepsilon_1$	-17.636	-17.583	-17.557	N/A
		$\Delta E$	12.708	12.694	12.660	N/A

## 4 Bulk calculations

### 4.1 Correlation between norm error and transferability

In order to confirm the correlation between the norm error and transferability, we plot the relative errors of the bulk properties of several elements with respect to the norm errors in Fig. S1. The norm error and each error of bulk properties are calculated as

$$\begin{aligned} \Delta N_{nc} &= \left[ \frac{\int_0^{r_c} (|\phi_0(r)|^2 - |\psi_0(r)|^2) dr}{\int_0^{r_c} |\psi_0(r)|^2 dr} + \frac{\int_0^{r_c} (|\phi_1(r)|^2 - |\psi_1(r)|^2) dr}{\int_0^{r_c} |\psi_1(r)|^2 dr} \right] \times \frac{100}{2}, \\ \Delta V_0 &= \frac{|V_0^{\text{NCPP}} - V_0^{\text{NNLP}}|}{V_0^{\text{NCPP}}} \times 100, \\ \Delta B_0 &= \frac{|B_0^{\text{NCPP}} - B_0^{\text{NNLP}}|}{B_0^{\text{NCPP}}} \times 100, \\ \Delta E_c &= \frac{|E_c^{\text{NCPP}} - E_c^{\text{NNLP}}|}{E_c^{\text{NCPP}}} \times 100. \end{aligned} \tag{1}$$

In eqn (1),  $\psi_l$  and  $\phi_l$  are the wavefunctions obtained from all-electron calculations and KS-DFT calculations with NNLP, respectively, and  $V_0$ ,  $B_0$ , and  $E_c$  are the equilibrium volumes, bulk modulus, and cohesive energy calculated by KS-DFT with NCPP and NNLP, respectively. The calculated systems are Li BCC, Be HCP, C diamond, Na BCC, Mg HCP, Al FCC, Si diamond, P orthorhombic, Ar FCC, K BCC, Ca FCC, Ga orthorhombic, Ge diamond, As trigonal, Se trigonal, Br orthorhombic, and Kr FCC.

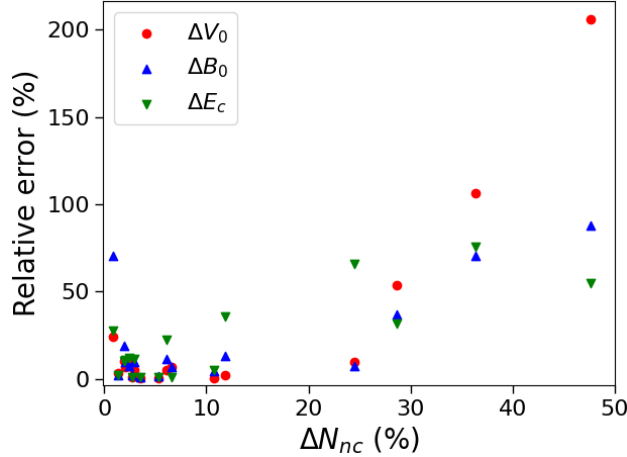


Figure S 1: The relative errors of the bulk properties of several elements with respect to the corresponding norm errors in atomic calculations. The calculated bulk properties are the equilibrium volume  $V_0$ , the bulk modulus  $B_0$ , and the cohesive energy  $E_c$ . The results were calculated by KS-DFT calculations with NCPP and NNLP. The results of NCPP were regarded as the reference. LDA was used as the exchange-correlation functional.

## 4.2 Band structure calculations

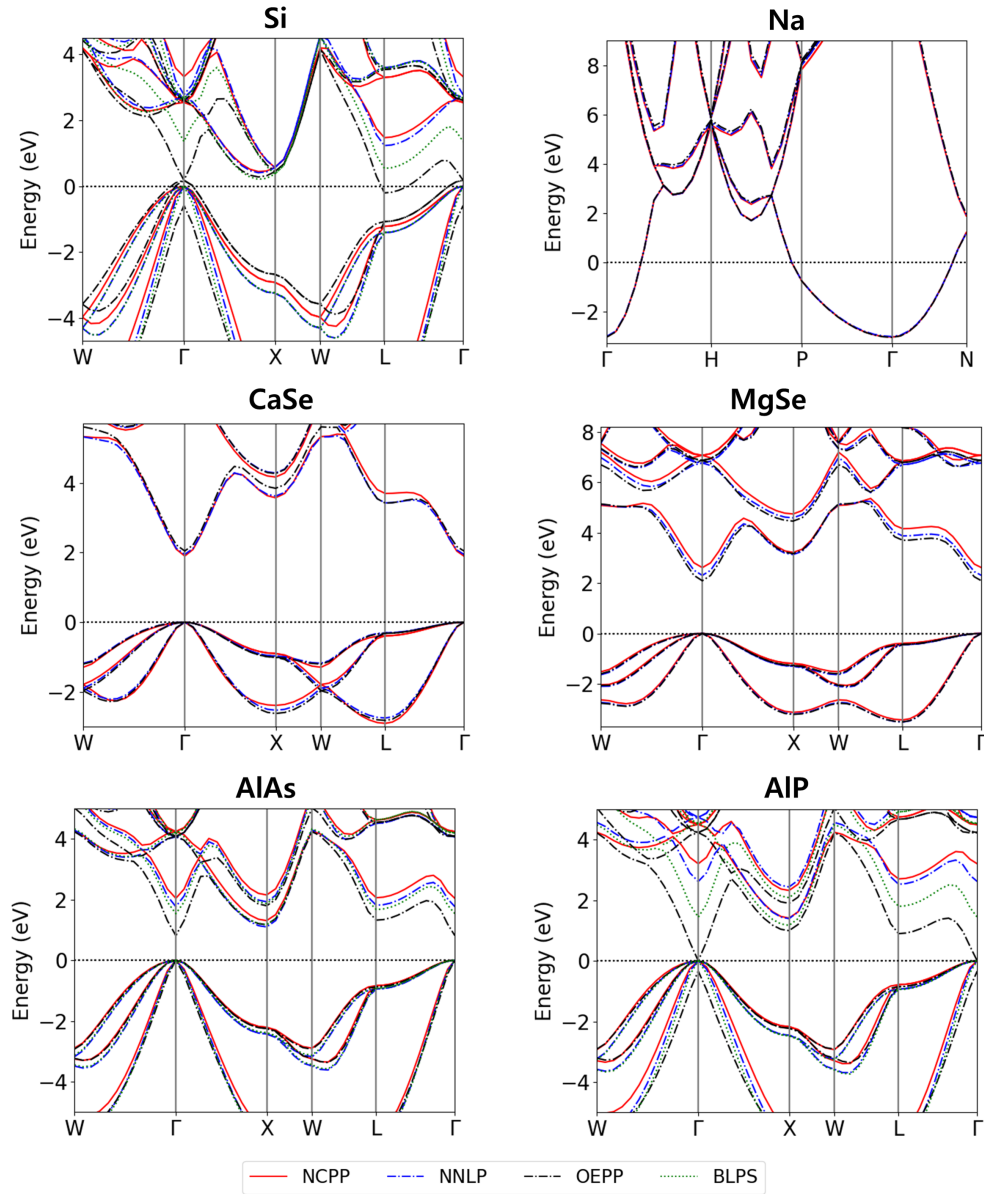


Figure S 2: The band structures of the Si diamond, Na BCC, CaSe RS, MgSe ZB, AlAs ZB, and AlP ZB calculated using KS-DFT with NCPP (red), NNLP (blue), OEPP (black), and BLPS (green). LDA was used as the exchange-correlation functional. The energy (y-axis) is the relative to the Fermi energy.



## 5 Effect of the cutoff radius on the accuracy in atomic calculations

To test the effect of the cutoff radius  $r_c$  on the accuracy in atomic calculations, we generated NNLPs for S with different  $r_c$  values (1.1959–2.1959 bohr at the interval of 0.1 bohr). All adjustable parameters to obtain the NNLPs were used as described in Section 1 except for  $r_c$  and  $w_2$ . Each  $w_2$  corresponding to the NNLP at the given  $r_c$  was optimized by the Bayesian optimization method. We performed atomic calculations using these NNLPs and calculated the errors in eigenvalues as defined in eq (12) in the main text.

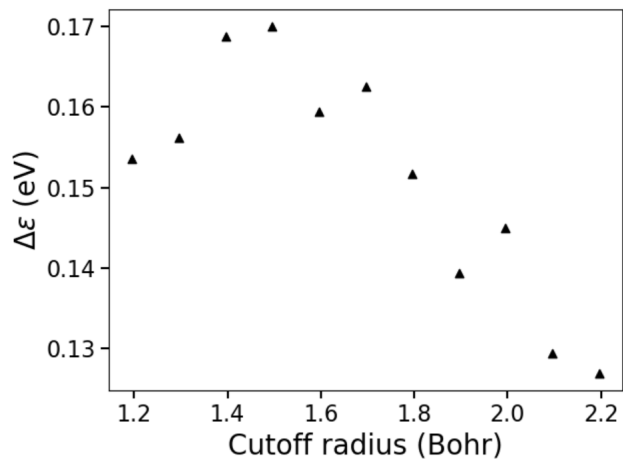


Figure S 3: The errors in eigenvalues as a function the cutoff radius. The results were calculated by KS-DFT with NNLPs of S generated with different cutoff radii. LDA was used as the exchange-correlation functional.