- Supporting Information -

First-Principles Calculations of Luminescence Spectra of Real-Scale Quantum Dots

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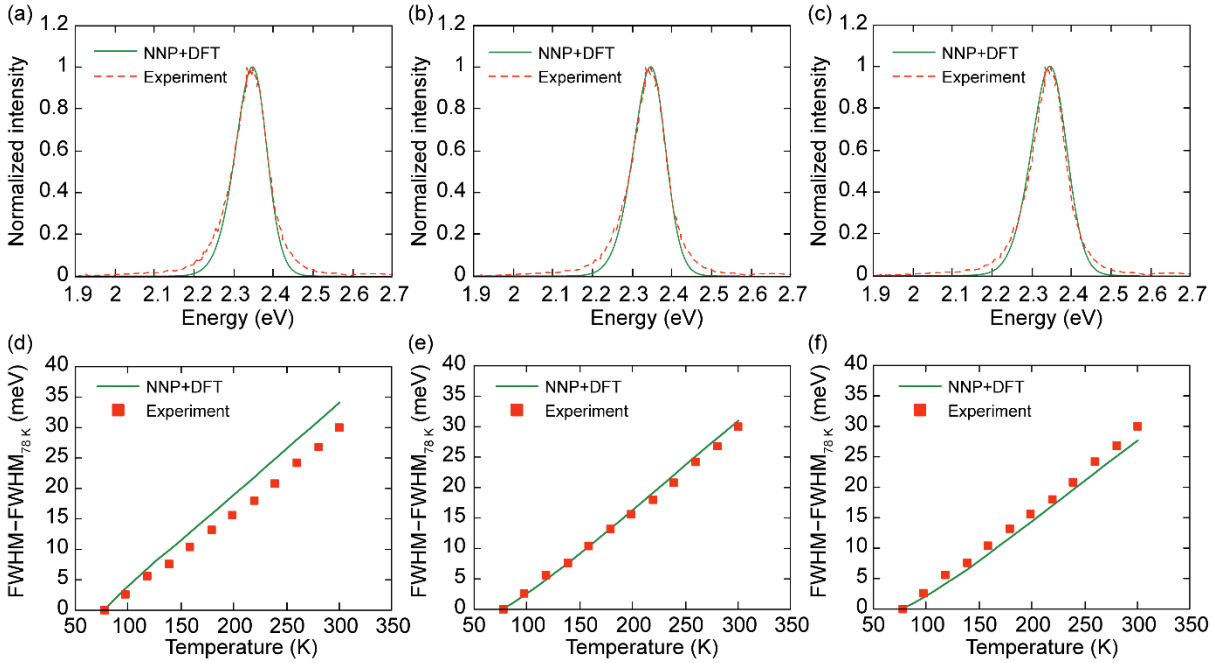

Figure S1. (a,b,c) Luminescence spectra and (d,e,f) temperature dependence of FWHM calculated using Gaussian linewidth of (5 meV, 10 meV, 20 meV).

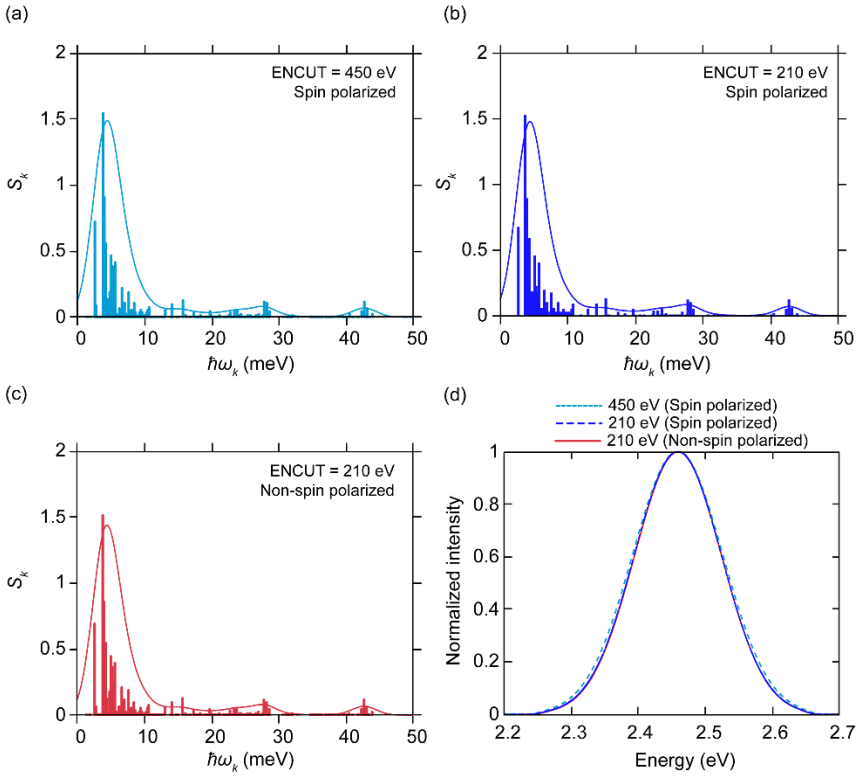

Figure S2. Partial HR factors of $(InP)_{22}/(ZnSe)_{73}/H_{110}$ calculated using cutoff energy of (a) 450 eV with spin-polarized calculation, (b) 210 with spin-polarized calculation and (c) 210 eV with non-spin-polarized calculation when calculating atomic forces. (d) The luminescence lineshapes calculated by three methods.

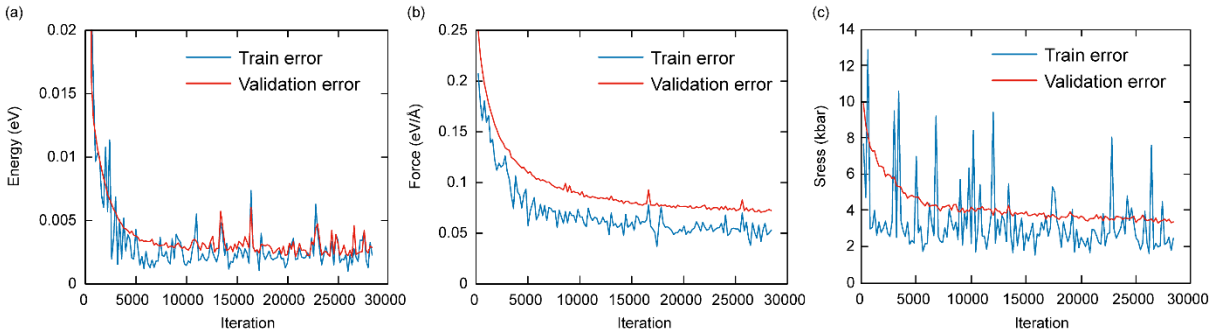

Figure S3. The learning curves of (a) energy, (b) force, and (c) stress errors. The larger fluctuation in the training error occurs because the number of reference structures considered for training NNP is relatively small (12 structures in a batch at a given iteration). Meanwhile, the validation set for evaluating the validation error includes even more structures (284 structures). The different behavior in the training and validation errors with respect to the training iteration is generally observed in NNP training using the batch-training method (for example, see Fig. S3 in ref. 19).

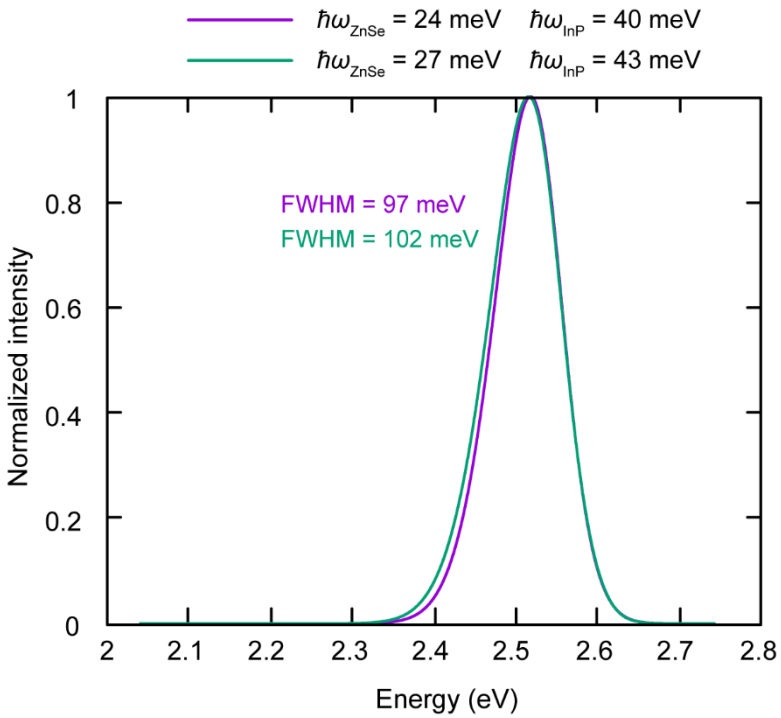

Figure S4. Luminescence spectra of $(InP)_{95}/(ZnSe)_{1283}/H_{650}$ QD calculated by effective EPC parameters. The purple line is the emission spectrum obtained using calculated optical-phonon frequencies and the green line is the emission spectrum obtained using experimental optical-phonon frequencies.

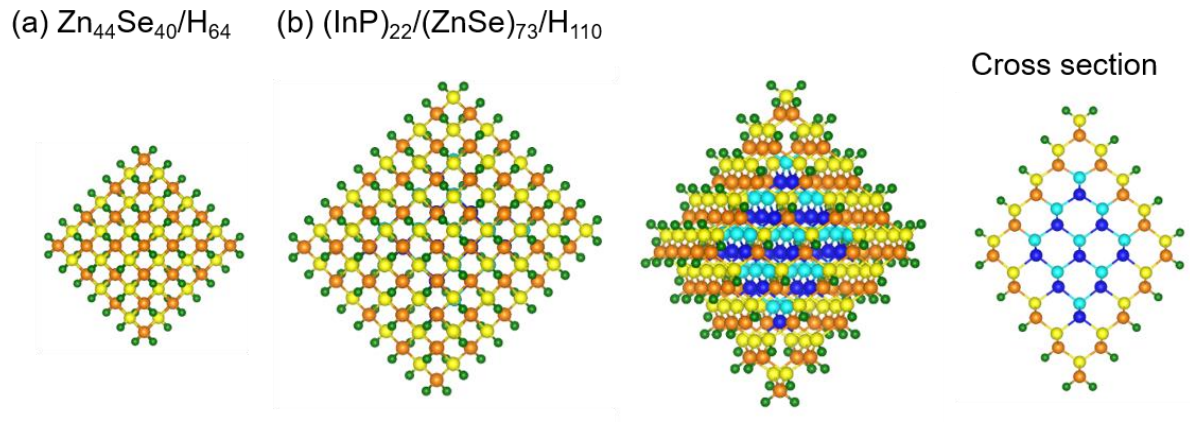

Figure S5. Atomic structures of (a) $Zn_{44}Se_{40}/H_{64}$ and (b) $(InP)_{22}/(ZnSe)_{73}/H_{110}$. See Figure 3 for details.

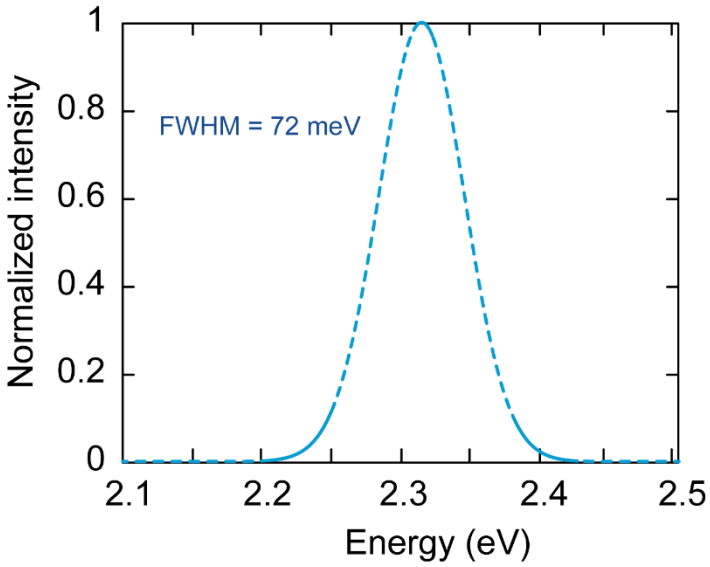

Figure S6. Luminescence lineshape of $(InP)_{95}/H_{110}$ QD.

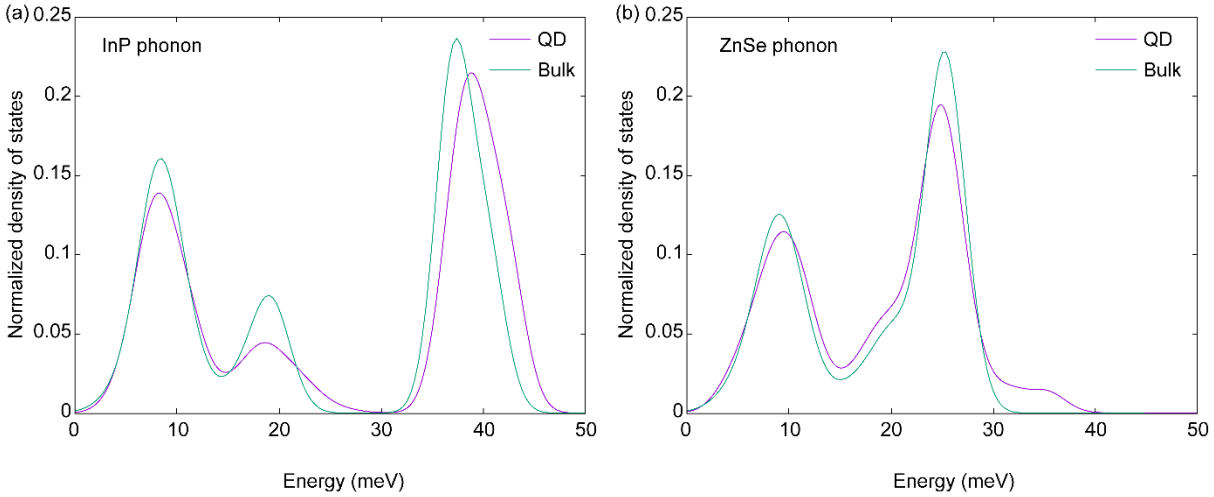

Figure S7. The density of states of bulk and QD phonons for (a) InP and (b) ZnSe. The QD data of each material was obtained by summation of contributions of the corresponding atoms to all of vibrational modes of a core-shell QD [$(InP)_{95}(ZnSe)_{1283}H_{650}$].

Table S1. Comparison of effective HR factors (S_{eff}) and FWHMs calculated using PBE+U and HSE06 functional of InP QD and ZnSe QD.

	Exchange-correlation functional	S_{eff}	FWHM (meV)
$In_{43}P_{44}H_{76}$ QD	PBE+U	2.60	97
	HSE06	3.00	94
$Zn_{43}Se_{44}H_{76}$ QD	PBE+U	4.08	100
	HSE06	3.20	109

Table S2. Lattice constants of InP and ZnSe.

	This work	Experiments [S5,S6]
InP	5.892 Å	5.869 Å
ZnSe	5.631 Å	5.650 Å
Lattice mismatch between InP and ZnSe	4.4%	3.7%

Table S3. Electron and hole effective masses of InP and ZnSe bulk.

		This work (m_e)	Experiment (m_e) [S1,S2]
InP	Electron	0.108	0.08
	Hole	0.108 (light hole) 0.822 (heavy hole)	0.121 (light hole) 0.531 (heavy hole)
ZnSe	Electron	0.154	0.17
	Hole	0.165 (light hole) 0.829 (heavy hole)	0.20 (light hole) 0.65 (heavy hole)

Table S4. Dielectric constants of InP and ZnSe bulk.

		This work	Experiment [S3,S4]
InP	Electronic part	10.23	9.52
	Ionic part	3.00	2.83
ZnSe	Electronic part	6.40	5.7
	Ionic part	2.95	2.9

Table S5. Summary of reference structures and root-mean-square erros (RMSEs) for validation set. See Figure 1 in the main text for detail atomic structures of reference structures.

Data description	# structures	# data points	MD temperature (K)	Energy RMSE (meV)	Force RMSE (meV/Å)	Stress RMSE (kbar)
InP MD	400	25600	1000 K	1.4	63.7	2.0
InP distorted crystal	61	3904	-	2.0	8.8	8.0
ZnSe MD	400	29504	1000 K	1.2	56.7	1.5
ZnSe distorted crystal	61	3904	-	2.0	2.3	1.5
ZnSe slab	100	12600	400 K	2.1	67.7	1.5
ZnSe/H QD	100	8600	300 K	2.3	131.7	0.3
InP/ZnSe interface	200	36400	1000 K	1.2	80.1	1.8
InP/ZnSe/H QD	100	23000	300 K	0.9	51.2	0.5
TOTAL	1422	143512	-	1.5	69.9	2.3

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