Supplemental Material:

Physical Origin of Color Changes in Lutetium Hydride under Pressure

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Table S1 lists the plasma frequencies and reflectivity of blue and red light of LuH_2 and LuH_3 under different pressures. Tables S2-S4 list the detailed structure information corresponding to Figs. S2-S4. Crystal structures corresponding to Figs. S5-S8 were previously suggested by Ref. [1]. Detailed structural information can be found in the supplementary materials of Ref. [1].

Figure S1 shows the real part of the dielectric functions of LuH_2 under different pressures. Figures S2-S8 show crystal structures and dielectric functions of N-doped lutetium hydrides calculated in this work. The contributions of interband and intraband transitions to dielectric functions at 0 GPa (at 25 GPa for Fig. S5) are plotted separately, and the evolution of the total dielectric function under different pressures is also shown. The reflectivity and color of those structures not displayed in the main text are shown here. Figures S9-S14 show the dielectric functions and reflectivity of LuH_2 and LuH_3 with different hydrogen vacancies ratios.

Figure S15 shows the calculated phonon dispersion of LuH_2 and LuH_3. Figs. S15 (a) and (b) show the harmonic phonon dispersion. It was calculated using the density-functional perturbation theory (DFPT) method, with phonon frequency determined via the PHONOPY code [2]. Figs. S15(c) and (d) show the anharmonic phonon dispersion. The anharmonic phonon frequency is calculated using the mode decomposition technique developed in DynaPhoPy [3]. This code uses the molecular dynamics trajectory calculated by VASP and projects it onto a set of harmonic phonon modes obtained by Phonopy. The power spectrum is then calculated using the maximum entropy method (MEM), and the peaks are fitted to Lorentzian functions to extract the anharmonic phonon properties.

Table S1. The plasma frequencies and reflectivity of typical blue (2.8 eV) R_{blue} and red (1.8 eV) R_{red} under different pressures.

Structure		LuH_2			LuH_3	
Pressure	ω_p (eV)	R_{blue}	R_{red}	ω_p (eV)	R_{blue}	R_{red}
0 GPa	5.82	0.45	0.28	1.62	0.42	0.41
10 GPa	6.00	0.34	0.25	2.18	0.42	0.41
20 GPa	6.11	0.29	0.48	2.36	0.44	0.41
30 GPa	6.17	0.27	0.57	2.89	0.44	0.42
40 GPa	6.25	0.25	0.60	2.74	0.43	0.44
50 GPa	6.33	0.23	0.60	2.22	0.44	0.43

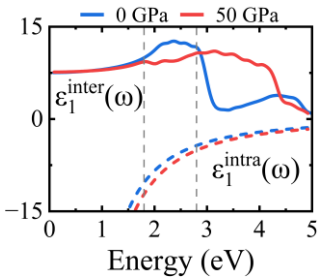

Fig. S1. The real part of the dielectric function contributed by interband (solid line) and intraband (dashed line) transitions.

Table S2. Structure information of $Lu_8H_{15}N$ ($LuH_{1.875}N_{0.125}$) ($a = b = c = 10.0660$ Å, $\alpha = \beta = \gamma = 90.00$ °).

#	atom	position		
1	Lu	0.3750	0.1250	0.1250
2	Lu	0.1250	-0.1250	0.1250
3	H	0.2500	0.0000	0.2500
4	H	0.0000	-0.2500	0.5000
5	H	0.0000	-0.5000	0.5000
6	H	0.0000	0.0000	0.5000
7	H	-0.2500	-0.7500	0.7500
8	N	-0.2500	-0.2500	0.7500

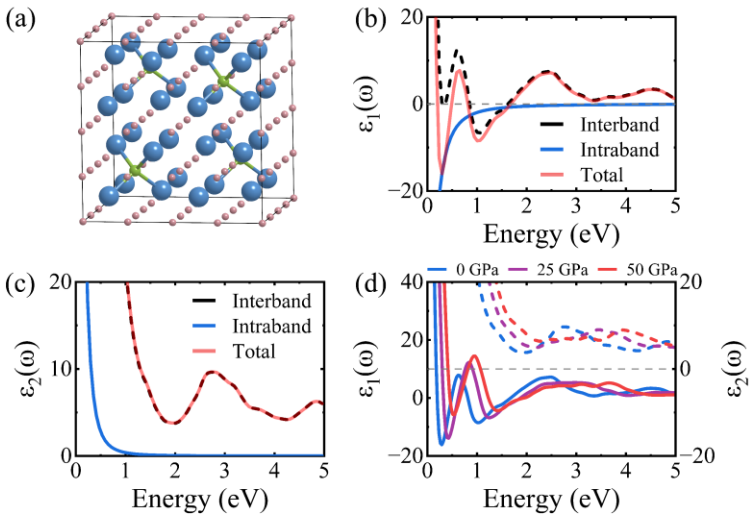

Fig. S2. (a) Crystal structure, (b)-(d) dielectric functions of $Lu_8H_{15}N$ ($LuH_{1.875}N_{0.125}$, nitrogen atom at the tetrahedral site). Solid lines and dashed lines in (d) represent the real part ε_1 and imaginary part ε_2 of the dielectric function, respectively.

Table S3. Structure information of $Lu_8H_{16}N$ ($LuH_2N_{0.125}$) ($a = b = c = 10.0660$ Å, $\alpha = \beta = \gamma = 90.00$ °).

#	atom	position		
1	Lu	-0.2500	0.2500	-0.2500
2	Lu	-0.7500	0.0000	0.0000
3	H	-0.3750	0.1250	-0.1250
4	H	-0.8750	-0.1250	0.1250
5	N	-0.5000	0.0000	0.0000

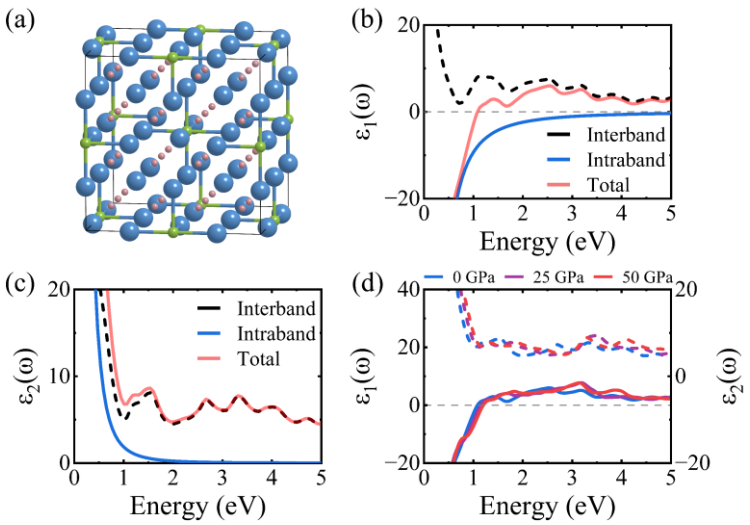

Fig. S3. (a) Crystal structure, (b)-(d) dielectric functions of $Lu_8H_{16}N$ ($LuH_2N_{0.125}$, nitrogen atom at the octahedral site).

Table S4. Structure information of $Lu_{18}H_{35}N$ ($LuH_{1.944}N_{0.056}$) ($a = 15.5127$ Å, $b = 10.6766$ Å, $c = 7.1177$ Å, $\alpha = \gamma = 90.00$ °, $\beta = 76.7376$ °).

#	atom	position		
1	Lu	-0.1667	0.1667	0.3333
2	Lu	-0.3333	0.3333	0.6667
3	Lu	-0.5000	0.1667	1.0000
4	Lu	-0.1667	0.1667	0.8333
5	Lu	-0.3333	0.3333	1.1667
6	Lu	-0.5000	0.1667	1.5000
7	Lu	0.0000	0.0000	0.0000
8	Lu	-0.3333	0.0000	0.6667
9	Lu	-0.6667	0.0000	1.3333
10	Lu	0.0000	0.0000	0.5000
11	Lu	-0.3333	0.0000	1.1667
12	Lu	-0.6667	0.0000	1.8333
13	H	-0.2500	0.1667	0.6250
14	H	-0.4167	0.1667	1.2083
15	H	-0.4167	0.3333	0.9583
16	H	-0.5833	0.3333	1.5417
17	H	-0.5833	0.1667	1.2917
18	H	-0.7500	0.1667	1.8750
19	H	-0.2500	0.1667	1.1250
20	H	-0.4167	0.1667	1.7083
21	H	-0.4167	0.3333	1.4583
22	H	-0.5833	0.3333	2.0417
23	H	-0.5833	0.1667	1.7917
24	H	-0.7500	0.1667	2.3750

25	H	-0.0833	0.0000	0.2917
26	H	-0.2500	0.0000	0.8750
27	H	-0.4167	0.0000	0.9583
28	H	-0.5833	0.0000	1.5417
29	H	-0.7500	0.0000	1.6250
30	H	-0.9167	0.0000	2.2083
31	H	-0.0833	0.0000	0.7917
32	H	-0.2500	0.0000	1.3750
33	H	-0.5833	0.0000	2.0417
34	H	-0.7500	0.0000	2.1250
35	H	-0.9167	0.0000	2.7083
36	N	-0.4167	0.0000	1.4583

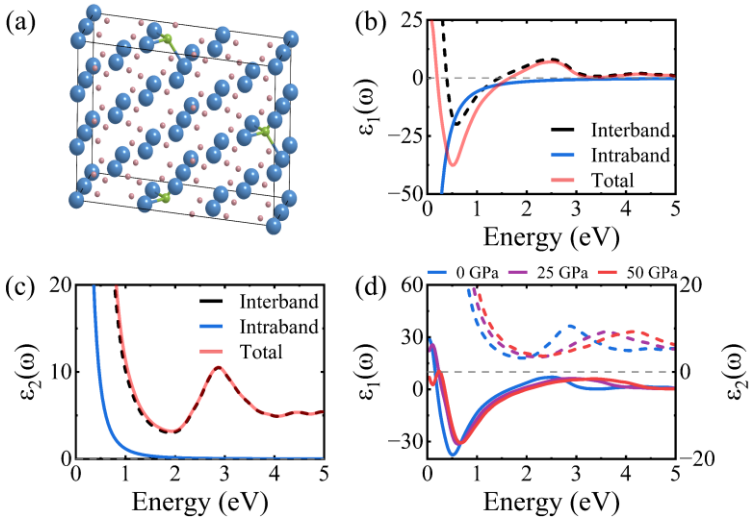

Fig. S4. (a) Crystal structure, (b)-(d) dielectric functions of $Lu_{18}H_{35}N$ ($LuH_{1.944}N_{0.056}$, nitrogen atom at the tetrahedral site).

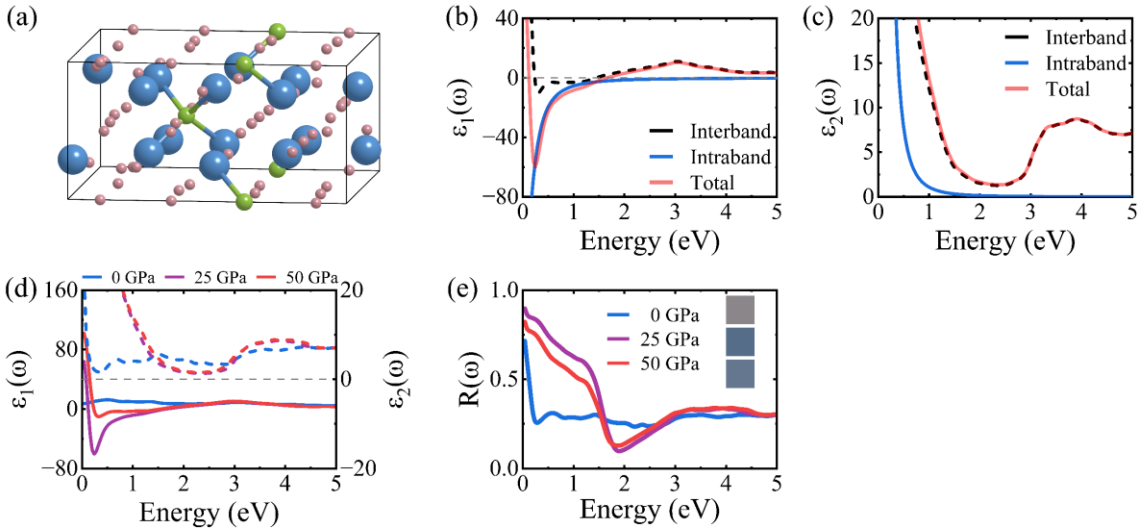

Fig. S5. (a) Crystal structure, (b)-(d) dielectric functions (at 25 GPa), (e) reflectivity and color of $Lu_8H_{23}N$ ($LuH_{2.875}N_{0.125}$, nitrogen atom at the tetrahedral site).

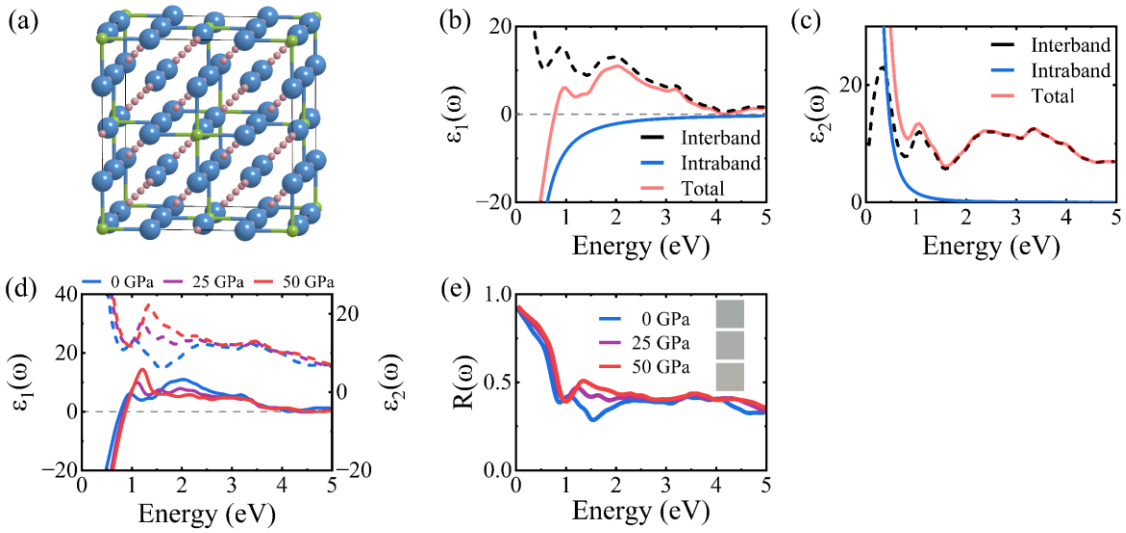

Fig. S6. (a) Crystal structure, (b)-(d) dielectric functions, (e) reflectivity and color of $Lu_8H_{21}N$ ($LuH_{2.625}N_{0.125}$, nitrogen atom at the octahedral site).

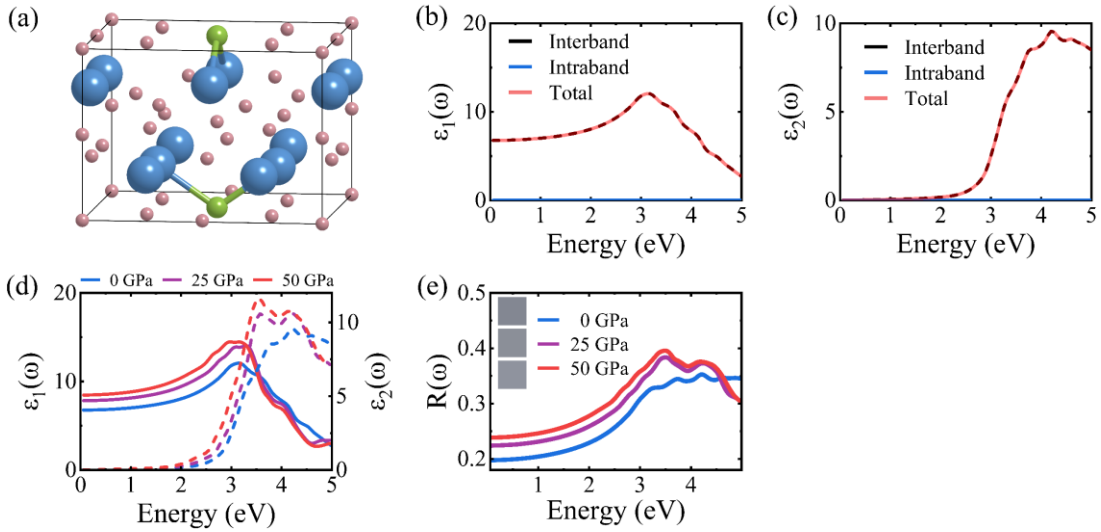

Fig. S7. (a) Crystal structure, (b)-(d) dielectric functions, (e) reflectivity and color of $Lu_8H_{21}N$ ($LuH_{2.625}N_{0.125}$, nitrogen atom at the tetrahedral site).

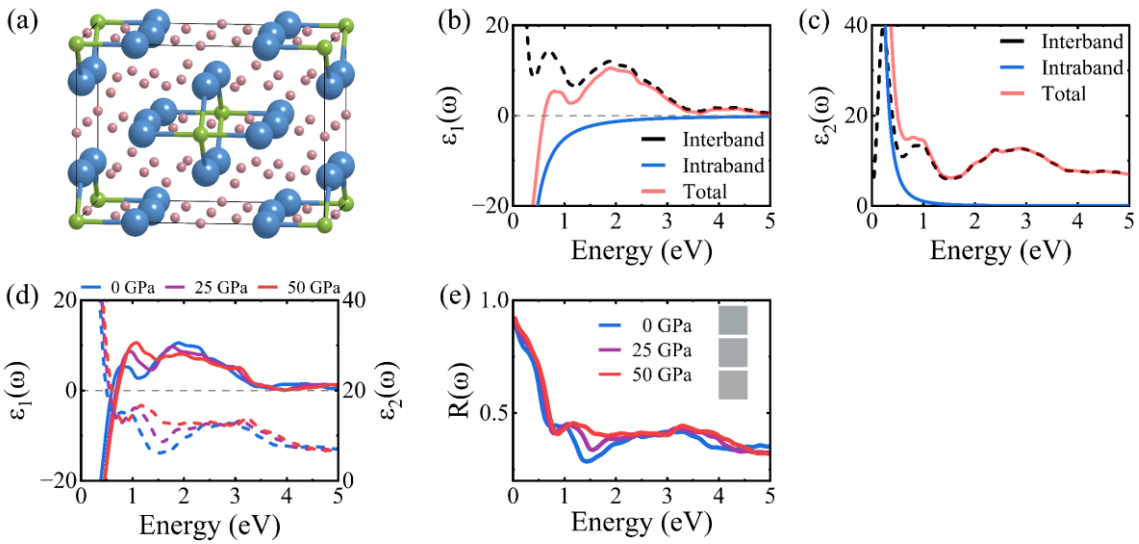

Fig. S8. (a) Crystal structure, (b)-(d) dielectric functions, (e) reflectivity and color of $Lu_8H_{23}N$ ($LuH_{2.875}N_{0.125}$, nitrogen atom at the octahedral site).

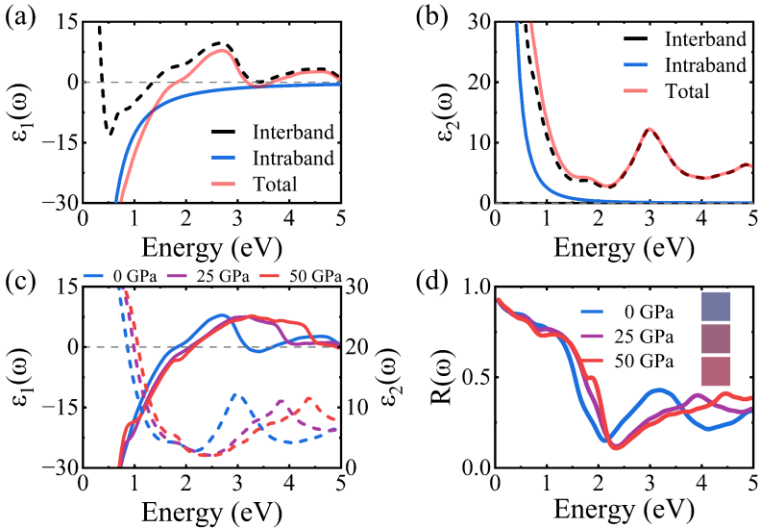

Fig. S9. Calculated real (a) and imaginary (b) parts of the dielectric function of $LuH_{1.875}$. (c) The dielectric function and (d) reflectivity under different pressures. The inset in (d) shows its color under corresponding pressure.

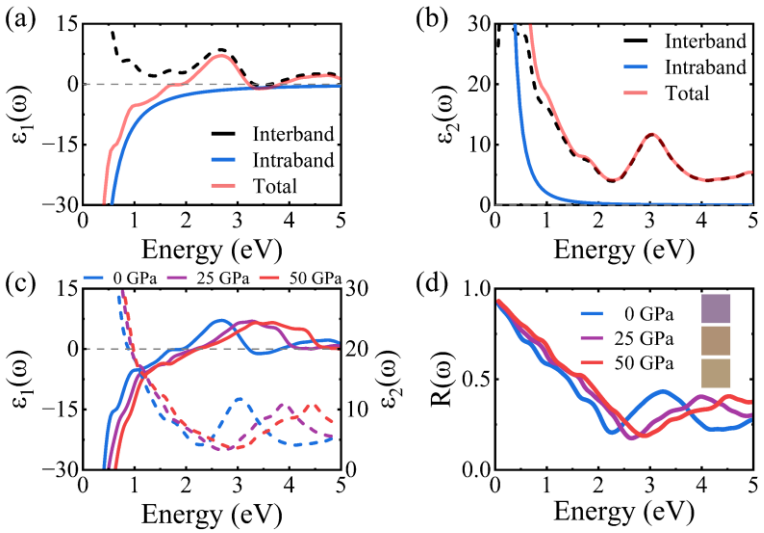

Fig. S10. Calculated real (a) and imaginary (b) parts of the dielectric function of $LuH_{1.750}$. (c) The dielectric function and (d) reflectivity under different pressures. The inset in (d) shows its color under corresponding pressure.

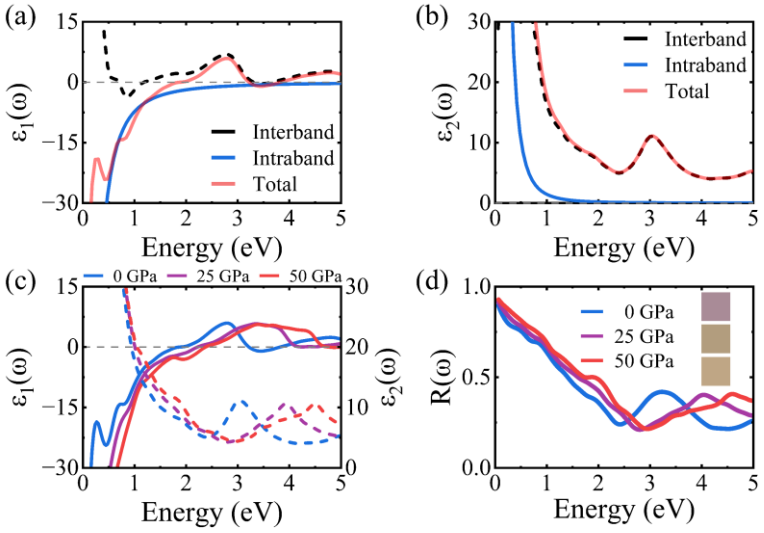

Fig. S11. Calculated real (a) and imaginary (b) parts of the dielectric function of $LuH_{1.625}$. (c) The dielectric function and (d) reflectivity under different pressures. The inset in (d) shows its color under corresponding pressure.

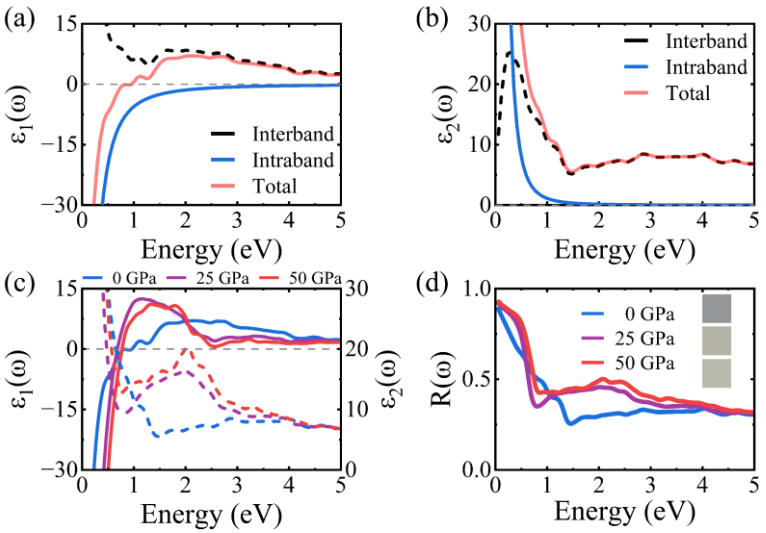

Fig. S12. Calculated real (a) and imaginary (b) parts of the dielectric function of $LuH_{2.875}$. (c) The dielectric function and (d) reflectivity under different pressures. The inset in (d) shows its color under corresponding pressure.

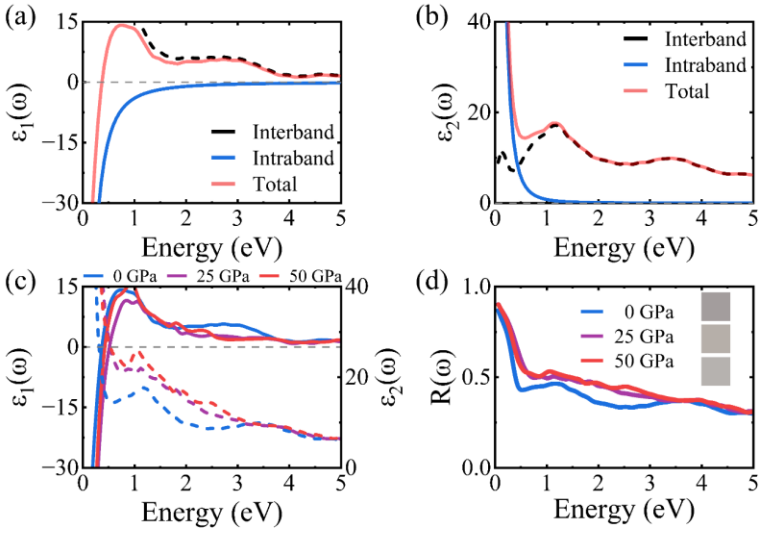

Fig. S13. Calculated real (a) and imaginary (b) parts of the dielectric function of $LuH_{2.750}$. (c) The dielectric function and (d) reflectivity under different pressures. The inset in (d) shows its color under corresponding pressure.

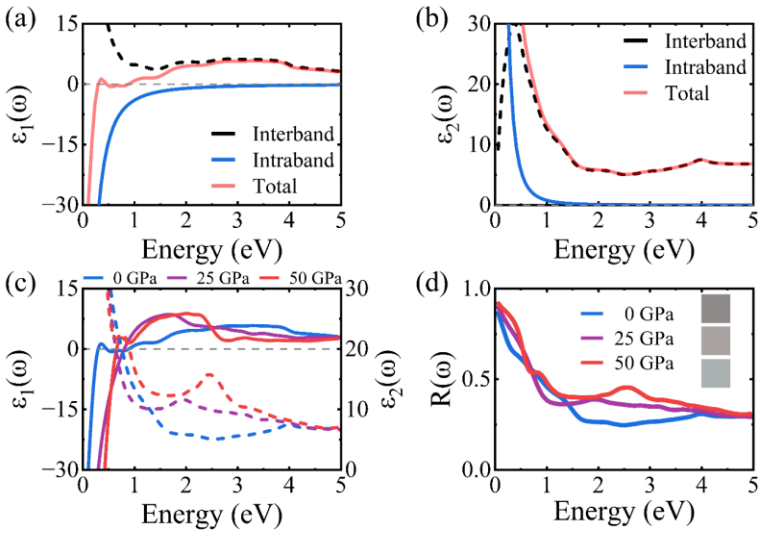

Fig. S14. Calculated real (a) and imaginary (b) parts of the dielectric function of $LuH_{2.625}$. (c) The dielectric function and (d) reflectivity under different pressures. The inset in (d) shows its color under corresponding pressure.

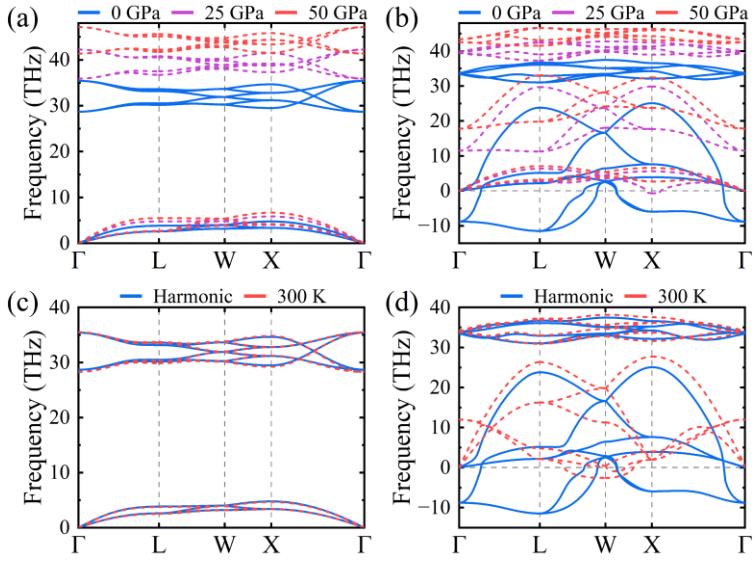

Fig. S15. Harmonic phonon dispersion of (a) LuH_2 and (b) LuH_3 under different pressures. Harmonic and anharmonic phonon dispersion at 300 K of (c) LuH_2 and (d) LuH_3.

References

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[2] Chaput L *et al.* 2011 *Phys. Rev. B* **84** 094302

[3] Carreras A, Togo A, Tanaka I 2017 *Comput. Phys. Commun.* **221** 221