Searching for Materials with High Refractive Index and Wide Band Gap: A First-Principles High-Throughput Study (Supplemental Material)

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I. HINTS ABOUT THE RELATIONSHIP BETWEEN ω_g AND E_g^d

The relationship between the average optical gap ω_g and the direct band gap E_g^d can formally be deduced from Eq. (A14) of the main text which only involves the JDOS $j(\omega)$. However, the form of $j(\omega)$ is often complex, which makes it impossible to solve the equation analytically.

Here, the solution is first derived for a simple model JDOS:

$$j(\omega) = \begin{cases} -\frac{6}{(2\sigma)^3} (\omega - E_g^d) \left(\omega - (E_g^d + 2\sigma) \right) & \text{for } E_g^d \le \omega \le E_g^d + 2\sigma \\ 0 & \text{elsewhere} \end{cases}$$
(S1)

This corresponds to a JDOS showing a single symmetric parabolic peak with a width σ (see Model 1 in Fig. S1(a)). The analytic solution of Eq. (A14) of the main text for this model JDOS is given by:

$$\omega_g^3 = -\frac{4}{3}\sigma^3 \left[\ln\left(\frac{E_g^d + 2\sigma}{E_g^d}\right) - \frac{2\sigma(E_g^d + \sigma)}{E_g^d(E_g^d + 2\sigma)} \right]^{-1}$$
(S2)



FIG. S1. (a) JDOS models (see text) and (b) corresponding relation between the average optical gap ω_g and ω_g - E_g^d (in eV) from Eq. (A14) of the main text. All models show the same trend which can be fitted using Eq. (S3). The results obtained by truncating the development to n=1 (as in Eq. (3) of the main text) are represented for Models 2, 3, and 4 using dotted, dashed, dot-dashed lines, respectively.

Using the properties of the logarithm and its Taylor expansion, we can finally write:

$$\omega_g = E_g^d + \sum_{n=0}^{\infty} \frac{c_n(\sigma)}{(E_g^d)^n} \tag{S3}$$

where the coefficients $c_n(\sigma)$ are functions of the width σ . By truncating the development to n=1, we find Eq. (4) of the main text.

Next, more complex JDOS models are considered. Model 2 consists of a skew normal distribution:

$$j(\omega) = \frac{A}{\sigma\sqrt{2\pi}} e^{-\frac{(\omega-\mu)^2}{2\sigma^2}} \left\{ 1 + \operatorname{erf}\left[\frac{\gamma(\omega-\mu)}{\sigma\sqrt{2}}\right] \right\}$$
(S4)

where A, μ , σ , and γ are the amplitude, the position, the width, and the skewness of the peak, respectively. Models 3 and 4 are obtained by summing two and three such skew normal distributions.

For all models, an analytic solution of Eq. (A14) of the main text is out of reach but ω_g can be computed numerically for different values of the peak position in order to determine its dependence with respect to E_g^d . In all cases, it is found that the solution has the form of Eq. (S3) and that its truncation to n=1 provides a good approximation (see Fig. S1(b)).



FIG. S2. Splitting of the $\omega_g \cdot E_g^d$ data points considering the different ranges of the width σ . For each panel, the dashed black line corresponds to $\omega_g = E_g^d + 6.85 - 1.68/E_g^d$ which was obtained by fitting all the data, while the colored lines are obtained considering only the data in the subset represented in the panel.

Furthermore, playing with the parameters, we confirm that, just like for Model 1, the coefficients $c_n(\sigma)$ in Eq. (S3) clearly depend on the width σ . These findings suggest that Eq. (S3) truncated to n=1 could be used for any JDOS. This is illustrated in Fig. S2 for all our calculated data. The dependence of the coefficients $c_n(\sigma)$ on the width σ has been highlighted by splitting the data into 6 groups according to the width of the real JDOS σ (computed as the difference between the mean value of the JDOS and the direct gap). This width depends on the dispersion of the bands, hence on the effective mass, and their distribution in energy, as illustrated in Fig. S3. This justifies the use of Eq. (4) in the main text.

II. COMPARISON OF THE n_s - E_g^d MODELS

As mentioned in the main text, different empirical or semi-empirical models have been proposed for the expected inverse relationship between the refractive index n_s and the direct band gap E_q^d . A review of such models was recently



FIG. S3. Schematic illustration of the dependence of the average optical gap ω_g on the width of the JDOS $j(\omega)$. Starting from a two-state system (a) with flat bands for which ω_g coincides with the direct band gap E_g^d and $j(\omega)$ is a Dirac peak, the graphs shows how ω_g is affected by (b) the dispersion of the bands which increases the width of the JDOS, (c) the band distribution in energy when new flat bands are added leading to new Dirac peaks in $j(\omega)$, and (d) the combination of both.

proposed by Tripathy [1]. In Fig. S4, we report the distributions of the absolute errors on the refractive index for various explicit functions of the direct band gap E_q^d compared to the DFPT computed data for the 4040 materials.



FIG. S4. Distribution of the absolute errors on the refractive index (for various explicit function of the direct band gap E_g^d vs. DFPT) for the 4040 materials. Our model (a) is compared to some well known empirical and semiempirical relations: (b) Ravindra *et al.* [2], (c) Moss [3], (d) Hervé and Vandamme [4], (d) Reddy and Anjaneyulu [5], and (e) Kumar and Singh [6]. For all the models, the mean absolute error (MAE), the 25th, 50th, 75th percentiles (P25, P50, and P75) are indicated.

We consider various descriptors of the distribution of the absolute errors: the mean absolute error (MAE), the 25th, 50th, 75th percentiles (P25, P50, and P75, respectively). All of them indicate that our new model describes the calculated DFPT data better than the previously proposed ones.

III. COMPARISON WITH THE PENN MODEL

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The model presented here can directly be connected to the so-called Penn model in which the static refractive index is given by the following equation:

$$n_s^2 = \varepsilon_{1s} = 1 + \left(\frac{\omega_p}{\omega_g}\right)^2,\tag{S5}$$

where ω_p is the Drude plasma frequency which is defined through the *f*-sum rule:

$$\int_0^\infty \omega \varepsilon_2(\omega) d\omega = \frac{\pi}{2} \omega_p^2.$$
(S6)

Introducing the imaginary part of the dielectric function given by Eq. (A10) of the main text into Eq. (S6) we then obtain:

$$\omega_p^2 = 8\pi K \int_0^\infty \frac{j(\omega)}{\omega} d\omega.$$
(S7)

If we now consider the two-state system introduced in the main text whose JDOS is given by $j(\omega) = J\delta(\omega - \omega_g)$, the resulting plasma frequency is simply given by:

$$\omega_p^2 = \frac{8\pi KJ}{\omega_g} = \frac{\omega_{\text{eff}}^3}{\omega_g}.$$
(S8)

The two expressions for the static refractive index given by Eq. (A15) of the main text and Eq. (S5) are thus strictly equivalent. Consequently, the model presented in this study is closely related to Penn model. Our model has, however, an important advantage over Penn model for analyzing the data in that the effective frequency ω_{eff} is clearly independent of the average optical gap ω_g (it only depends on the integral of the JDOS J and the average transition probability K), while the plasma frequency ω_p is not.

IV. AVERAGE TRANSITION PROBABILITY VS. INTEGRAL OF THE JDOS



FIG. S5. Calculated values of the average transition probability K and integral of the JDOS J computed up to a frequency ω_{max} for the complete dataset of materials. The data are reported as solid circles the color of which refers to the value of ω_{eff} as indicated in the color bar.

From Eq. (A13) of the main manuscript we have that ω_{eff} is related to the product of the average transition probability K and the integral of the JDOS J. In Fig. S5, we show the distribution of all our data points as a function of these two quantities (shown in a logarithmic scale for sake of clarity). The data points have been colored according to ω_{eff} . Though K is smaller than 1 for the vast majority of materials, both K and J impact the value of ω_{eff} .

V. ANALYSIS BY CLASS OF COMPOUNDS

In order to analyze the trend of the data in the different compounds, we separated the oxides (3375 out of 4040) in four classes (see main text): TMOs with empty d shell (1st group) (671 materials); TMOs with partially filled d shell (2nd group) (303); main-group oxides (1520 materials); and lanthanide oxides (747). The result of our analysis is shown in Fig. S6. For each class, an ellipse (in black in the figure) is defined as follows. Its center is located at the average value of E_g^d and n_s for the corresponding distribution. The orientation and lengths of its axes are determined using principal component analysis for the materials which belong to the region with a density larger than 75%.



FIG. S6. Static refractive index n_s as a function of the direct band gap E_g^d for the 4 classes of materials: (a) TMOs with empty d shell (1st group) in red, (b) TMOs with partially filled d shell (2nd group) in blue, (c) main-group oxides in green, and (d) lanthanide oxides in orange. The solid line corresponds to Eq. (16) of the main text for the average value of $\omega_{\text{eff}}=10.75$ eV. For each class, the probability density function is computed in the distribution for the refractive index as a function of the band gap via a Kernel-Density Estimation (KDE) using a Gaussian kernel. The darker regions for each class represent the higher value for the distribution density.

VI. TABLES OF COMPOUNDS

In this section, we provide various tables with the 10 materials with the highest refractive index for a given direct band gap range. In each table the chemical formula, MP identification (mp-id), average refractive index (n_s) , diagonal components of the refractive index tensor (n_1, n_2, n_3) , direct band gap in eV (E_g^d) , the effective frequency in eV (ω_{eff}) , the average optical gap in eV (ω_g) and the average effective mass of the transitions μ are shown. The compounds are sorted by the value of the average refractive index. The full list containing the 4040 compounds taken in consideration in this study can be found in the comma-separated values (CSV) file (db.csv).

VII. IMPORTANCE OF THE EXCHANGE-CORRELATION FUNCTIONAL

It is clear that the exchange-correlation (XC) functional will affect the electronic structure (the band gap and, possibly, the band dispersion) and the optical properties. Here, we analyze how using HSE instead of PBE changes the calculated value of ω_g for the top materials in the Tables S1-S6. To this end, in Fig. S7, we compare the JDOS $j(\omega)$ and $j(\omega)/\omega^3$ computed with computed with both XC functionals. Obviously, the PBE gaps are smaller than the HSE ones so a scissor Δ was applied to the PBE electronic structure in order to match the HSE gap. A comparison of the average gap ω_g computed with PBE+ Δ_{HSE} and HSE for the selected materials is also shown in Table S7. For each material the value of the Δ_{HSE} scissor operator is reported. Looking at the different ω_g values one can see that there is a small difference considering the two XC functionals, with an absolute error ranging from 0.15 to 1.09 eV.

Formula	mp-id	n_s	n_1	n_2	n_3	E_g^d	$\omega_{ m eff}$	ω_g	μ
TePb	mp-19717	5.12	5.12	5.12	5.12	0.87	17.47	5.96	0.08
FeS_2	mp-1522	4.59	4.41	4.77	4.60	1.17	11.88	4.37	0.83
$\mathrm{Tl}_2\mathrm{S}$	mp-667	4.46	4.48	4.58	4.32	0.86	19.12	7.18	0.25
SnSe	mp-691	4.25	4.60	4.25	3.91	0.59	7.62	2.95	0.24
RuS_2	mp-2030	4.09	4.09	4.09	4.09	0.91	13.35	5.32	0.34
$\mathrm{Sb}_2\mathrm{Se}_3$	mp-2160	3.90	4.30	4.28	3.10	0.80	10.76	4.45	0.47
GeSe	mp-700	3.79	3.97	3.78	3.62	0.93	13.97	5.89	0.33
${\rm Te}_2{\rm Mo}$	mp-602	3.69	4.28	4.28	2.53	1.07	10.09	4.32	0.48
SnS	mp-2231	3.68	3.85	3.58	3.60	1.07	15.37	6.61	0.24
$\mathrm{Sm}_{2}\mathrm{Te}_{3}$	mp-684911	3.68	3.68	3.68	3.67	0.99	9.84	4.23	0.32

TABLE S1. List of compounds with $0.0 \le E_g^d \le 2.0 \ eV$.

TABLE S2. List of compounds with $2.0 \le E_g^d \le 3.0 \ eV$.

Formula	mp-id	n_s	n_1	n_2	n_3	E_g^d	$\omega_{ m eff}$	ω_g	μ
TiO_2	mp-34688	2.61	2.65	2.65	2.52	2.35	10.77	5.99	0.94
$YbTe_2$	mp-1779	2.60	2.60	2.60	2.60	2.46	12.98	7.23	0.19
$\mathrm{Bi}_{2}\mathrm{O}_{3}$	mp-23262	2.57	2.61	2.63	2.48	2.46	12.79	7.19	0.58
${ m Ti}_3{ m PbO}_7$	mp-504427	2.57	2.60	2.59	2.51	2.32	14.01	7.90	114.11
CaTe	mp-1519	2.57	2.57	2.57	2.57	2.62	11.96	6.74	0.23
$\mathrm{LiBi}_3\mathrm{I}_2\mathrm{O}_4$	mp-775430	2.56	2.62	2.62	2.44	2.25	12.40	7.00	0.55
B_8O	mp-530033	2.56	2.55	2.57	2.56	2.46	18.29	10.33	1.11
GeI_2	mp-27922	2.56	2.80	2.80	2.07	2.42	11.67	6.59	1.58
$\mathrm{Bi}_4\mathrm{I}_2\mathrm{O}_5$	mp-30130	2.55	2.59	2.52	2.55	2.37	11.74	6.65	0.97
TiPbO_3	mp-20459	2.54	2.57	2.57	2.47	2.80	14.44	8.21	0.68

TABLE S3. List of compounds with $3.0 \le E_g^d \le 4.0 \ eV$.

Formula	mp-id	n_s	n_1	n_2	n_3	E_g^d	$\omega_{ m eff}$	ω_g	μ
ZrSO	mp-3519	2.78	2.78	2.78	2.78	3.01	12.71	6.73	0.70
HfSO	mp-7787	2.65	2.65	2.65	2.65	3.56	13.22	7.25	0.70
${\rm LiNb_3O_8}$	mp-3368	2.39	2.45	2.38	2.34	3.00	12.15	7.25	3.62
$\mathrm{Ta_2Pb_2O_7}$	mp-755663	2.39	2.40	2.37	2.39	3.11	15.36	9.17	5.62
Nb_2ZnO_6	mp-17177	2.38	2.48	2.32	2.34	3.30	13.40	8.02	3.25
$LaTa_7O_{19}$	mp-14485	2.38	2.35	2.35	2.43	3.13	13.36	8.00	2.68
BiClO	mp-22939	2.37	2.50	2.50	2.10	3.00	13.45	8.07	0.48
$NdTa_7O_{19}$	mp-14676	2.37	2.34	2.34	2.42	3.17	13.91	8.35	3.26
YTa_7O_{19}	mp-772036	2.36	2.34	2.34	2.42	3.16	13.81	8.31	3.45
BaTiO_3	mp-5020	2.35	2.35	2.35	2.35	2.97	12.74	7.69	1.11

TABLE S4. List of compounds with $4.0 \le E_g^d \le 5.0 \ eV$.

Formula	mp-id	n_s	n_1	n_2	n_3	E_g^d	$\omega_{ m eff}$	ω_g	μ
ZrO_2	mp-755089	2.26	2.28	2.20	2.30	4.15	13.78	8.59	1.35
$ScTaO_4$	mp-558781	2.25	2.27	2.30	2.20	4.03	13.28	8.31	1.61
HfO_2	mp-1858	2.24	2.22	2.24	2.27	4.41	15.02	9.44	1.22
$PrScO_3$	mp-559756	2.22	2.24	2.23	2.19	4.26	14.05	8.90	1.15
$\mathrm{Sr}_{2}\mathrm{Zr}_{7}\mathrm{O}_{16}$	mp-770419	2.22	2.20	2.20	2.25	4.02	13.42	8.51	3.67
NdScO_3	mp-31117	2.21	2.24	2.22	2.18	4.29	13.97	8.88	1.24
ThO_2	mp-643	2.20	2.20	2.20	2.20	4.51	13.40	8.56	0.83
HfO_2	mp-775757	2.20	2.23	2.21	2.16	4.02	14.84	9.48	1.44
SmScO_3	mp-31118	2.20	2.22	2.21	2.16	4.36	13.78	8.81	1.41
HfO_{2}	mp-352	2.19	2.22	2.21	2.14	4.10	14.85	9.51	1.76

TABLE S5. List of compounds with $5.0 \le E_g^d \le 6.0 \ eV$.

Formula	mp-id	n_s	n_1	n_2	n_3	E_g^d	$\omega_{ m eff}$	ω_g	μ
BeS	mp-422	2.29	2.29	2.29	2.29	5.63	18.11	11.17	0.24
BeSiN_2	mp-7913	2.13	2.16	2.14	2.11	5.30	18.77	12.30	0.39
${\rm LiSi_2N_3}$	mp-5853	2.07	2.09	2.05	2.08	5.48	17.50	11.77	0.42
DyClO	mp-755323	2.07	2.09	2.09	2.02	5.17	14.68	9.88	0.45
HoClO	mp-29731	2.07	2.09	2.09	2.02	5.18	14.10	9.50	0.61
HfSiO_4	mp-4609	1.98	1.97	1.99	1.98	5.65	15.77	11.03	2.10
ErBO_3	mp-10791	1.95	1.94	1.96	1.94	5.15	15.93	11.32	2.68
$\mathrm{Pr}_3\mathrm{Si}_2\mathrm{ClO}_8$	mp-554826	1.93	1.93	1.93	1.93	5.15	14.79	10.56	1.25
$\mathrm{Nd}_2\mathrm{Be}_2\mathrm{SiO}_7$	mp-9077	1.92	1.89	1.93	1.93	5.03	15.86	11.41	0.64
$Y_2Be_2SiO_7$	mp-6655	1.90	1.87	1.91	1.91	5.19	15.57	11.31	0.63

TABLE S6. List of compounds with $6.0 \le E_g^d \le 7.0 \ eV$.

Formula	mp-id	n_s	n_1	n_2	n_3	E_g^d	$\omega_{ m eff}$	ω_g	μ
$BeAl_2O_4$	mp-3081	1.78	1.78	1.78	1.78	6.12	18.71	14.45	0.37
$MgAlBO_4$	mp-8376	1.72	1.72	1.72	1.72	6.23	17.76	14.20	0.42
LiCl	mp-22905	1.72	1.72	1.72	1.72	6.25	15.92	12.75	0.38
LaF_3	mp-905	1.70	1.70	1.70	1.70	6.04	10.20	8.24	9.19
$\mathrm{NaPr_2S_2O_8F_3}$	mp-560673	1.70	1.70	1.71	1.68	6.18	14.97	12.13	6.05
$\mathrm{CaB}_{2}\mathrm{O}_{4}$	mp-8056	1.69	1.73	1.71	1.64	6.09	15.18	12.31	0.60
$Al_6B_5O_15F_3$	mp-6738	1.68	1.68	1.68	1.67	6.17	17.08	14.03	0.53
$BaBePO_4F$	mp-754604	1.67	1.67	1.67	1.67	6.24	15.22	12.52	0.47
SiO_2	mp-549166	1.65	1.82	1.57	1.57	6.00	16.26	13.53	0.47
$\rm LiB_3O_5$	mp-3660	1.62	1.63	1.64	1.60	6.35	16.13	13.70	1.02

TABLE S7. Comparison of the ω_g values (in eV) computed considering PBE+ Δ_{HSE} and HSE for the top materials in the Tables S1-S6. For each material also the value of the scissor operator Δ_{HSE} (in eV) is reported.

Formula	MP-id	$\Delta_{ m HSE}$	$\omega_g \ (\text{PBE} + \Delta_{\text{HSE}})$	ω_g (HSE)
TePb	mp-19717	0.54	7.09	6.00
TiO_2	mp-34688	1.57	7.73	8.00
ZrSO	mp-3519	1.30	8.24	8.39
$\rm ZrO_2$	mp-755089	1.84	10.52	10.75
BeS	mp-422	1.26	12.41	12.64
$BeAl_2O_4$	mp-3081	2.25	16.59	16.79



FIG. S7. Optical functions $j(\omega)$ and $j(\omega)/\omega^3$ computed with PBE+ Δ_{HSE} and HSE for the top materials in the Tables S1-S6.

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