Expanding the Photonic Palette: Exploring High Index Materials

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ABSTRACT: While the photonic community is occupied with exotic concepts portending a grand future and fame if not fortune, I respectfully entertain the possibility that a humble concept of simply increasing the refractive index by a modest factor may have a far greater payoff in many walks of life. With that in mind, I explore why higher index materials have not yet materialized and point out a few tentative directions for the search of these elusive materials, be they natural or artificial.

KEYWORDS: *refractive index, optical materials, metamaterials*
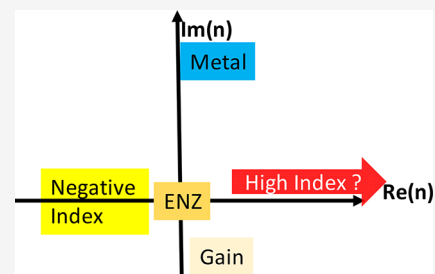
In every technology, material properties play a paramount role, and among those properties, one can pinpoint a few that ultimately enable a wide variety of devices and techniques. For electronics, such key material properties are conductivity, carrier mobility, and dielectric constant; for thermal control, it is thermal conductivity and specific heat; for micromechanics, it is Young's modulus, hardness, and elasticity; for acoustics, it is the speed of sound and so on. What is common among all of these properties is that Nature and human determination have given us an extremely wide range of materials where the aforementioned (and many other) properties vary by many orders of magnitude. This provides engineers and designers with a very broad design space that is continuously expanding as new materials enter the mainstream. However, the situation is quite different in photonics where the defining material property is the refractive index. Here, the refractive index n determines the minimum achievable features in imaging and lithography through the diffraction limit, λ/n, while the index contrast between two materials determines the minimum size of the integrated optical components and the optical strength of components, such as lenses, diffraction elements, and meta-surfaces.

Although the index can be as high as 3.5 in the near-IR (Si) and 5.8 in the mid-IR (PbTe), given its importance, it is astonishing that since the days of Carl Zeiss the range of refractive indices in high-quality optical materials widely available in the visible range has been limited to 1.38 (MgF_2) to 2.4 (TiO_2), as one can see from Figure 1.[1]

It would be expected that, with all the revolutionary developments in materials science that took hold in the last decades, a substantial effort would be directed at expanding the limited palette of refractive indices available to photonics practitioners. Yet, clearly this does not appear to be the case. Instead, through the last couple of decades, the photonics

community has been on a wild goose chase for far more exotic materials, with a negative[2,3] or near-zero[4] index, topological materials,[5] and ubiquitous 2D materials,[6] with graphene[7] being the king of them all. A true deluge of titles in reputable publications has materialized, loaded with superlatives like "Giant", "Super", "Colossal", "Ultra", "Extraordinary", "Exceptional", "Perfect", and so on. The pay-off so far has been rather modest, with no practical low loss negative and near-zero index materials emerging[8−10] and topological materials (in my view) being more of a solution in search of a problem. As far as two-dimensional materials are concerned, while they may exhibit interesting effects per unit thickness, all their purported advantages happen to be in vain, as more than a single atomic layer is required for any optical phenomenon (other than absorption) to have appreciable efficiency,[11] sufficient to be presented on a graph with a vertical scale featuring some meaningful units rather than ubiquitous "*arb. u*". One can also add to the above list the infatuation with metals (plasmonics[12]) even when faced with the undeniable fact that, unlike electronics, where amplification is easily available, photonics is far less tolerant of the loss and the loss in metals is inherently huge.[13] Due to this and also limits imposed by kinetic inductance,[14] plasmonics has not found too many applications in optical range beyond sensing[15,16] and the newly emerging science of single photon emission[17] (whose ultimate value hinges upon the eventual spread of quantum information
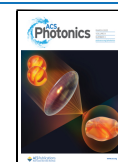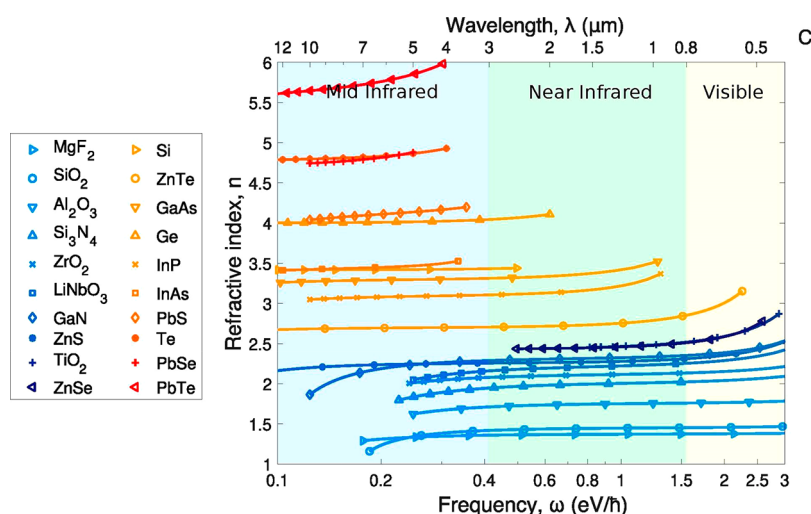
Figure 1. Indices of refraction for widely available materials. Adapted with permission from ref 1. Copyright 2020 The Optical Society.

processing, a topic on which I do not dare to speculate as, regrettably, I do not expect to witness it in my lifetime[18]). Lest it create an impression of me unjustly criticizing the community, here is my mea culpa: I have been just as guilty of participating in the mad scramble for the next fashionable material du jour as anyone else, publishing extensively in many of the aforementioned fields, although perhaps trying to retain some vestiges of sober judgment and not get carried away with my own rhetoric. Obviously, I am not the first one to raise these questions and many honest and fair (and, hence, unpopular) critical assessments of recently or currently "hot" topics can be found in the literature.[9,10,18−21]

Moving from this constructive criticism to a more positive discussion, it is hard to avoid the topic of metamaterials,[22] a term, which these days encompasses just about anything that can be conceivably fabricated (or envisioned) with diverse small scale techniques, be it lithography, nanoassembly, or epitaxial growth. While great progress in fabrication[23] and modeling[24,25] has been achieved in pursuit of three-dimensional optical metamaterials, overall they have not lived up to their early promise, but at the same time their two-dimensional offspring, metasurfaces,[26] have withstood the reality test and already are rapidly becoming an integral part of photonic techniques and devices. The original ideas for metasurfaces all relied on using metals,[27] but the metal metastructures for the optical range have not advanced far beyond the publication in reputable journal stage (not to say that this stage is not critically important in assuring an uninterrupted supply of scientific manpower and funding). The reasons are multiple, but one can relate them all to a hardly groundbreaking declaration, just as it is problematic to cook up a delicious dish from bad ingredients, it is also hard to cobble together a high quality optical metamaterial (surface) from low optical quality materials, such as metals with their high loss. At the same time, all-dielectric metasurfaces,[28−30] using true and tried high quality and low loss materials did take off and have shown performance metrics that are equal or even surpassing those of conventional optics.[29,31] But note that a large, and perhaps defining, factor in metasurface success has been the incorporation of high index material, TiO_2, with an index of 2.4, in contrast to the conventional optics relying on SiO_2 (n = 1.45). With optical power proportional to $n − 1$, it is no wonder that meta lenses can be three times thinner than silica optics for comparable performance! Similarly, in the near-IR

region, Si (n = 3.4) on SiO_2 metasurfaces exhibit enviable performance characteristics. And yet, even with these highly available indices, most metasurfaces fabricated today do not look at all like a flat prairie landscape as it was envisioned at their conception. In fact, under an electronic microscope, they look more like the island of Manhattan, with tall and narrow pillars of skyscrapers arranged in the square blocks, with high aspect ratios[32] required to attain phase and group delays needed to achieve a decent performance. In the absence of higher index materials, one is forced to further increase the height of metasurfaces creating fabrication difficulties.

Therefore, it is not really a stretch to imagine that, if even higher refractive indices were available to the designers, the performance of metasurfaces would reach heights that today appear unattainable. Through the years, the argument for discovering high index materials has been rather narrowly focused on one obvious goal; with a higher index, one can increase the resolution of imaging optics, reduce the weight and thickness of lenses, and also facilitate aberration correction. Now, however, the advent of metasurfaces, photonic crystals, and photonic integrated circuits[33] (PICs) makes the quest for high index materials if anything more urgent and important as performance in all these techniques is hinged upon high index contrast. With materials having a higher refractive index (while obviously maintaining transparency), one can envision PICs with smaller footprint devices, narrow low-loss waveguides with tighter bending radii,[34] photonic crystals that exhibit wider bandgaps, large numerical aperture metasurfaces, high resolution immersion optics, sensors,[16] solar cells where higher index means more light being trapped inside,[35] and many other devices. I would even venture to mention how high index materials may benefit such esoteric concepts as optical cloaking and transformational optics,[36] but prefer not to be carried away, leaving these enticing topics to more daring souls, and concentrate on earthlier concepts.

To summarize, perhaps the time has come for us to take a fresh look at the issue that has been considered settled since around the 19th century, if not earlier; namely, why are the values of refractive indices what they are, and what is it that prevents us from getting the larger values? In this essay, I embark on this less trodden path. First, I remind the readers about the origin of the refractive index in condensed matter, emphasizing the fact that unlike absorption properties that are determined

mostly by the states near the lowest bandgap, refraction is determined by the states throughout the entire Brillouin zone. It makes an estimate both more difficult and, at the same time, easy if one makes a few realistic approximations. Then, a simple polarizable bond model[37] will be reviewed, the estimate of the refractive index will be made, and the limitations will naturally emerge. Following that, the different means to achieve a high index combined with low loss in natural and artificial materials will be considered, with guarded optimism expressed, stating that 50−100% enhancement of the refractive index across a relatively narrow band is not entirely out of reach if a concerted effort is mounted. While not meriting such aforementioned superlatives as "giant" and "ultra-broad-band", the prospects of higher density PICs, more powerful and versatile metalenses, and a factor of 2 improvement of resolution in immersion optics should hopefully make this paper worthy of reading beyond this point.

■ WHY IS THE REFRACTIVE INDEX RANGE SO LIMITED?

Although it is universally known that the refractive indices ranges are quite small, the reason is perhaps less well recognized. On the most basic level it follows directly from the classical Lorentz model in which a polarizable electron cloud is kept in its place by a Coulomb attraction to one (as in the case of isolated atoms) or two or more ions in the case of polarizable bonds in molecules and condensed matter. In the most simple textbook example, the electron cloud is spherical with the "atomic" radius r_a, as shown in Figure 2a.
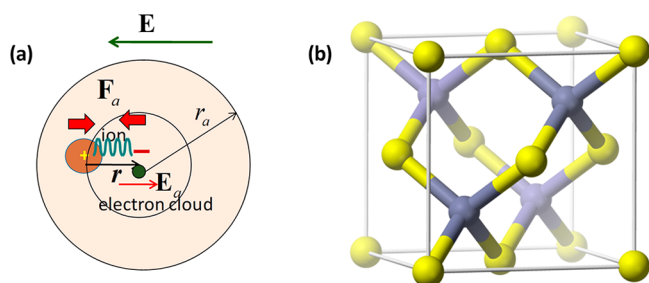

Figure 2. (a) Lorentz model used in the classical derivation of dielectric permittivity. **E** is the external field and $\mathbf{E}_a$ is the intrinsic field of the electron cloud. (b) Tetrahedral bonds in a diamond or zinc blend lattice.

From trivial electrostatic considerations, the restoring force acting on an electron that is moved by distance **r** from the equilibrium position is the same as the one acting on the ion

$$\mathbf{F}_a = -\frac{e^2}{4\pi\varepsilon_0 r_a^3}\mathbf{r} \equiv -K\mathbf{r} \tag{1}$$

where K can be thought of as a "spring coefficient". The resonant frequency of the Lorentz oscillator is $\omega_0 = \sqrt{K/m_0}$, and the dielectric constant can be found (neglecting damping) as

$$\varepsilon_r = 1 + \chi_{\text{ion}} + \frac{\omega_p^2}{\omega_0^2 - \omega^2} \tag{2}$$

where χ_{ion} is the ionic susceptibility, while the plasma frequency is $\omega_p = \sqrt{Ne^2/\varepsilon_0 m_0}$, and N is the density of the polarizable electrons. For frequencies well below ω_0 (i.e., in the trans-

parency region), but well above the resonant frequencies of the ionic vibrations (which are typically less than 10 THz), one can approximate $\varepsilon_r \sim 1 + \omega_p^2/\omega_0^2$. Substituting ω_0 and ω_p, we immediately obtain

$$\varepsilon_r(0) \approx 1 + \frac{Ne^2}{m_0\varepsilon_0}\left(\frac{e^2}{4\pi\varepsilon_0 r_a^3 m_0}\right)^{-1} = 1 + 4\pi Nr_a^3 \sim 1 + 3f_e \tag{3}$$

where $f_e = \frac{4}{3}\pi r_a^3 N$ is a fraction of the volume occupied by the electron cloud. Considering that, according to Pauli's principle, two electrons with opposite spins can occupy the same space, one can assume that $0 < f_e < 2$, which immediately presents us with a range of possible refractive indices of between 1 and 2.6, and that is right where the experimental values in the visible range are. Obviously, in the materials that are transparent only in the IR range, the refractive index increases to a higher value, which may be construed as the increased overlap of the electron orbitals (i.e., the effective size of wave function exceeding the distance between the ions) associated with the material becoming more metallic.[38]

In general, there exists an empirical Moss Rule[39] connecting the off-resonant refractive index and energy gap

$$n^4 E_G \approx 95 \text{ eV} \tag{4}$$

Although relatively primitive, the picture presented so far very well describes the limits on the refractive indexes imposed by Nature. Indeed, the authors of a remarkable recent work[40] have performed an exhaustive search of the data on the refractive index and its dispersion $dn/d\omega$ that confirmed eq 2. Furthermore, they have shown (based on the oscillator sum rule and the Kramer Kronig relation) that since, in agreement with eq 1, the only feasible way of enhancing the refractive index is to come close to the resonance, a strict constraint is placed on the refractive index and its dispersion,

$$n \leq \left(\frac{\omega_p^2}{\omega}\frac{dn}{d\omega}\right)^{1/3} \tag{5}$$

So, the possibility of getting a broadband large refractive index appears to be quite remote, but for many modern applications broadband operation is not necessary. Furthermore, large chromatic dispersion often comes in handy for aberration corrections, especially in hybrid lens/metasurface systems. Then, for a moment, keeping the Lorentzian character of eq 1 and assuming the fwhm of absorption $\Delta\omega$, we can obtain a maximum permittivity of

$$\varepsilon_r(\omega_0 - \Delta\omega/2) \approx 1 + \chi_{\text{ion}} + \frac{\omega_p^2}{\omega_0^2}Q \tag{6}$$

where $Q = \omega_0/\Delta\omega$. Therefore, the resonant refractive index $n_{\text{res}} \approx n(0)Q^{1/2}$ can be enhanced by a factor of 2−3, assuming that Q can be as high as 10, and a visible refractive index of 5−7 would have a transformative impact in photonics. On its surface, $Q \geq 10$ looks quite realistic; indeed, even much narrower optical transitions are routinely observed in gases and in materials doped with, for instance, rare earth ions. But, once the density of the active atoms/ions/molecules increases, which is necessary for a high index, harsh reality sets in as discrete energy levels broaden into energy bands, to the degree that for most solid materials Q only slightly exceeds unity; hence, the resonant enhancement of the refractive index is, at best, 30−40% for ionic materials like LiF, where the refractive index is small anyway, and
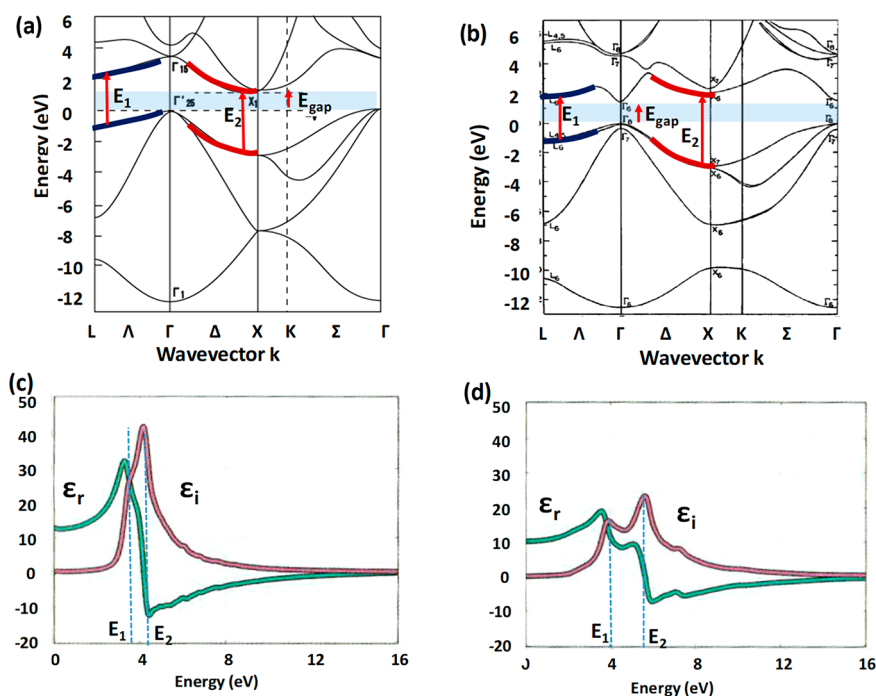
Figure 3. Band structures of (a) Si and (b) GaAs with the highlighted states that are the most responsible for the refractive index.[51] Real and imaginary permittivities of (c) Si and (d) GaAs. Note that the real part of permittivity does shave negative values, but unfortunately only in the high loss region.

far less for high index covalent materials, like a diamond, where the resonant enhancement is only about 10%.

To understand how broadening affects the refractive index, a more refined approach is needed. For that we shall revert to the bond theory of dielectric response,[37,41−45] developed more than half a century ago, that is, when all the towering figures of that period were busy producing hype-free gems of papers in *Physical Review*, as glossier publications were yet to be born, and the term "high impact" was primarily invoked in the context of lethal or near-lethal car wrecks.

A MORE REFINED LOOK AT THE PROBLEM

Leaving this nostalgic digression aside, according to the aforementioned works, the dielectric constant in the optical range is related to bond polarizabilities in the material. The occupied (unoccupied) bond states $u_v(\mathbf{r})$ ($u_c(\mathbf{r})$) have bonding (antibonding) character and correspond to valence (conduction) bands. Bond polarizabilities are defined by the transition dipole $e\mathbf{d}_{vc} = e\langle u_v|\mathbf{r}|u_c\rangle$, as shown in Figure 2b for the case of zinc blende or wurtzite lattice, characteristic of many covalent materials, such as II−VI and III−V, as well as group IV (in which case it is called a diamond lattice). In each case there are four hybrid orbitals in a tetrahedral arrangement surrounding each atom, as shown in Figure 2.

Summing over all the bonds, one can obtain the expression for the dielectric constant

$$\varepsilon_r = 1 + \chi_{\text{ion}} + \frac{2E_{vc}Ne^2d_{cv}^2/\varepsilon_0}{E_{vc}^2 - (\hbar\omega)^2} \tag{7}$$

where E_{vc} is the transition energy of the two states. Introducing the oscillator strength as

$$f_{cv} = 2m_0 E_{vc}d_{cv}^2/\hbar^2 \tag{8}$$

we easily obtain the same eq 2 modified by the oscillator strength,

$$\varepsilon_r = 1 + \chi_{\text{ion}} + \frac{f_{cv}\omega_{\text{P}}^2}{\omega_{cv}^2 - \omega^2} \tag{9}$$

For the strong allowed transition, f_{cv} approaches unity, and essentially the same limitations to the refractive index apply.

The bonds, however, are broadened into the bands, each state being a function of wave vector $\mathbf{k}$ and written as $\Psi_n(\mathbf{k}) = u_n(\mathbf{k})\exp(i\mathbf{k}\cdot\mathbf{r})$, where $u_n(\mathbf{k})$ is the periodic Bloch function, and index n stands for the conduction (c) or valence (v) band. Therefore, the dielectric constant has to be evaluated as a sum over all the possible transitions between the states in two bands

$$\varepsilon_r(\omega) = 1 + \chi_{\text{ion}} + \frac{e^2}{\varepsilon_0 V} \sum_k \frac{2E_{vc}(\mathbf{k})d_{cv}^2(\mathbf{k})}{E_{vc}^2(\mathbf{k}) - (\hbar\omega)^2} \tag{10}$$

where V is the volume, and also included is the imaginary part (neglecting off-resonant absorption by ions)

$$\varepsilon_i(\omega) = \frac{\pi e^2}{\varepsilon_0 V} \sum_k d_{cv}^2(\mathbf{k})\delta(E_{vc}(\mathbf{k}) - \hbar\omega) \tag{11}$$

Strictly speaking, the dipole moment is not easily defined in the extended periodic system,[46] and for calculating the resonant phenomena, such as the absorption spectrum, (eq 11) one typically uses the matrix element of momentum,[47] $\mathbf{p}_{vc} = i\hbar\langle u_v|\nabla|u_c\rangle$. However, if one introduces $\mathbf{d}_{vc}(\mathbf{k}) = \mathbf{p}_{vc}(\mathbf{k})/m\omega_{vc}(\mathbf{k})$, for the calculations of the off-resonant phenomena, such as refractive index, then the coordinate rather than the momentum gauge becomes preferable, because one can truncate the summation in (eq 10) at high energies.[48,49]

Let us now take a look at the characteristic band structures of Si and GaAs shown in Figure 3a and b, respectively. At first glance, the picture looks messy and bears no resemblance to the bond orbital picture of Figure 2b. At closer look, one realizes that in the 3D Brillouin zone (BZ) most of the states are located close to the zone edges, that is, X and L points. The states near the
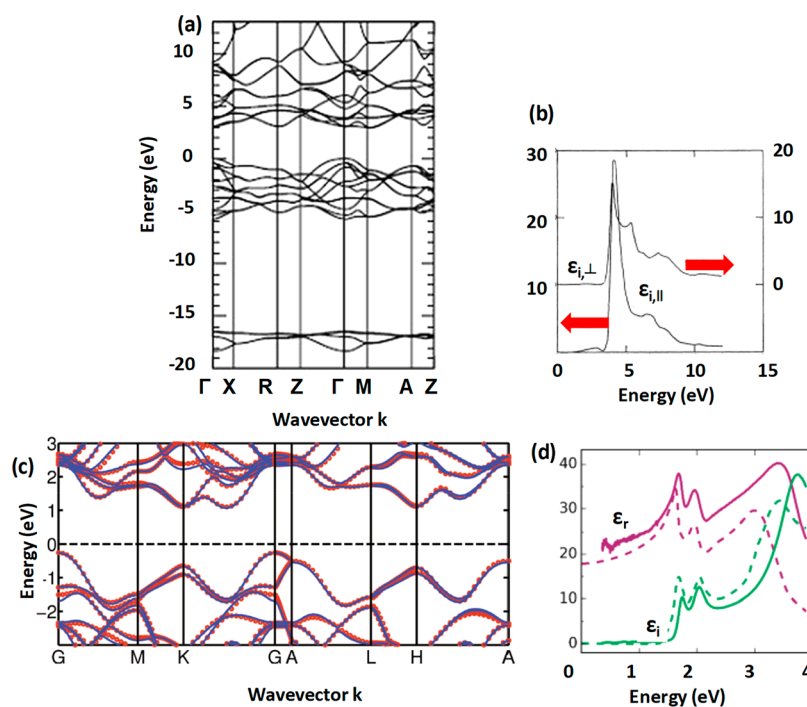
Figure 4. (a) Band structure of TiO$_2$ (rutile).[54] (b) Imaginary part of TiO$_2$ permittivity normal and parallel to the axis.[45] (c) Band structure of bulk MoS$_2$.[60] (d) Real and imaginary parts of the permittivity of MoS$_2$ for the monolayer (solid curves) and bulk (dashed curves).[61]

conduction and valence band extrema, separated by the bandgap energy E_{gap}, may play a central role in determining absorption and emission properties (as well as electronic properties in doped materials), but their contribution to the dielectric constant and refractive index is negligibly small, simply because there are so few of them. That is why the refractive indices of direct (GaAs) and indirect (Si) semiconductors are remarkably similar in the transparency region, as shown in Figure 3c,d, where the spectra of real and imaginary parts of permittivity are plotted.

One can also notice that along the directions pointing away from the center of BZ, the CB and VB run parallel to each other,[37] as highlighted in Figure 3a,b. Therefore, large numbers of transitions have energy close to E_1 and E_2, and the spectrum of ε_i, that is, absorption, shows distinct peaks at those energies, and, accordingly, the real part of ε experiences a Lorentz-like dispersion in the vicinity of these frequencies, becoming negative beyond E_2. Clearly, these resonances are nothing but the original resonances associated with tetrahedral hybrid orbitals broadened by the coupling between the individual orbitals. And this fact, of course, presents one with an opportunity to represent the dielectric constant (at least in the transparency region) well above the phonon resonances as

$$\varepsilon_r \approx 1 + \frac{\hbar^2 \omega_p^2}{E_P^2 - \hbar^2 \omega^2} \tag{12}$$

where E_P is called the Penn gap[44] that is typically between E_1 and E_2, closer to E_2.[50] For example, for Si, $E_P \approx 4.8$ eV, and for GaAs, $E_P \approx 5.1$ eV. One may think of the Penn gap as some average transition energy in which most of the oscillator strength is concentrated and which defines the refractive index, while the fundamental bandgap E_g defines the absorption properties.

Here one should point out the key difference between the Lorentz model and the actual solid state. In the Lorentz model, residual absorption far away from resonance decays slowly as

$\Delta \omega \omega / (\omega_0^2 - \omega^2)^2$, and therefore, it is always present at optical frequencies. But, the Lorentz model fails far from the resonance if there are no real states enabling the transition with the conservation of energy. In the solid state, transitions below the bandgap energy are possible due to either impurities or phonons, and the absorption decreases exponentially as $\exp[-(E_{gap} - \hbar \omega)/E_0]$ — Urbach tail,[52] where for high quality single crystal materials, E_0 is typically a few tens of meV. (An alternative sub-bandgap absorption mechanism due to excitonic transitions is discussed farther on.) Therefore, once the photon energy is roughly 100 meV below the bandgap, the absorption becomes negligibly small.

The strategy in the quest for a high index material is then to identify the materials in which the difference between the effective resonance energy E_P, and the absorption edge E_{gap} is smallest and obtains the resonantly enhanced index

$$n_{max} \sim \sqrt{1 + \frac{\hbar \omega_p^2}{E_P^2 - E_{gap}^2}} \tag{13}$$

Now, in this simple picture, one can discern why only a very small enhancement of the refractive index below the absorption edge can be observed with most of the covalent materials. With $E_{gap} = 1.1$ eV $\approx E_P/4.3$ for Si and $E_{gap} = 1.35$ eV $\approx E_P/3.8$ for GaAs, one only expects a moderate change in the refractive index by 3−4% relative to very long wavelengths, as one can indeed observe in Figure 1. Indeed, for most tetrahedral materials, the value of $E_P - E_{gap}$ is nearly constant and ranges between 3.5 and 4.5 eV,[53] indicating that resonant enhancement of refractive index is unattainable in these materials.

■ WHAT CAN BE DONE ABOUT IT?

So, let us convey in a simple form the dilemma facing us: to obtain a large index, a large number of large dipoles must be densely packed into a unit volume of material, which of course

contains a built-in self-contradiction. As if it were not enough, they have to be packed in such a way that the transition does not broaden into wide bands. Let us see what, if anything, can be done about it?

Narrow valence and conduction bands are typically formed by *d* shells of atoms, rather than *s* and *p* shells of covalent materials. The best known example is familiar TiO_2 (rutile, whose band structure is shown in Figure 4a), in which the VB is composed primarily of oxygen p-orbitals, while the CB consists of Ti 3d states;[54] hence, the peak of absorption, as shown in Figure 4b, occurs at a critical point of 4 eV,[45] while the band edge is around $E_{gap} \sim 3.3$ eV. Therefore, according to (eq 12), one expects a relatively large increase in the refractive index near the resonance. Indeed, in bulk rutile, the refractive index[55] increases from 2.4 at 1.5 μm to 2.9 at 0.4 μm, more than a 20% enhancement. In thin film[56] TiO_2, the refractive index increases from 2.05 to 2.52, an even larger enhancement near the absorption edge.

One should note that other materials where bonds are at least partially made up by *d*-shells of transition metals, such as $LiNbO_3$, $SrTiO_3$,[57] and other inorganic perovskites, also have large indices and a very strong dispersion near the absorption edge.[58] This fact shows that *d*-orbitals may be key to getting a high refractive index at least in a relatively narrow spectral region. In that respect, recently discovered all-inorganic metal halide perovskites may carry a promise.[59]

Another approach to the reduction of the width of the band is to consider materials in which some of the bonds have a weak, van der Waals (VdW) character, such as transition metal dichalcogenides (TMDC), WSe_2, MoS_2, and others.[62] In these materials, covalent bonds lying in a plane are highly polarizable, while the interlayer coupling is weak and does not contribute much to band broadening. The CB and VB in TMDCs have largely the character of *d*-shells of transition metals;[60] hence, the bands are relatively flat (as seen in Figure 4c). Also, these materials show strong excitonic transitions. Three factors affecting the index, namely, VdW bonding, d-shell character of conduction bands, and strong exciton transition result in bulk TMDCs exhibiting high indices of 3.6 for W-based and 4.4 for Mo-based[61,63−65] (Figure 4d), while being transparent all the way to 600 and 1000 nm, respectively, which means that these materials outperform TiO_2 and Si. Since optical power scales with $n - 1$, one can see that MoS_2 offers at least a 35% improvement in thickness relative to Si, while for the PICs, where integration density depends on the evanescent wave in the cladding, improvement can be even higher. In fact, TMDC application to metasurfaces[66] and PICs[67] have already been implemented. The fact that in TMDC the refractive index is mostly enhanced only for polarization normal to the axis[68] does not impede these applications, since PICs typically operate with one (TE in this case) polarization, while in the meta lenses the light is polarized normal to the optical axis.

So, the recipe for achieving high index is simply to try to operate in the vicinity of a relatively narrow resonance. Such resonances seem to appear stronger in VdW materials due to three factors mentioned above (d shells, VdW bond, exciton), and since only a relatively small fraction of these families has been explored, it is not unfathomable that some material may show a refractive index of 5−6 in the near-IR and may be 4 through the entire visible range. Since monolayers of the VdW materials tend to have a wider bandgap than bulk, one may consider heterostructures in which single layers of VdW materials are separated by wide bandgap spacers, like BN;[69]

this way, a somewhat smaller index can be traded for a wider transmission window.

On a more futuristic level, the writing is on the wall; the way to increase the index is to somehow develop a stoichiometric high density arrangement of atoms with strong and narrow transitions. That would require the active atoms to be widely separated, causing reduction in density and, hence, plasma frequency. But the reduction in the width of the band, determined by the overlap of wave functions on different atomic sites, would be exponential, and therefore, overall, the resonant refractive index would be high. Indeed, according to eq 6, if the atomic spacing is increased by a factor of 2 from 1.5 to 3 Å, ω_p^2 gets smaller by a factor of 8, while the bandwidth, according to a simple tight binding calculation, gets reduced from a few eV to less than a hundred meV to provide a strong resonant enhancement of the index. The question is how to keep the separation between ions to such large distances, clearly exceeding the bond lengths in any material (the scale of the bond length is dictated by the Bohr radius). Perhaps one can use lattices with underoxidized alkali-earth metal ions such as Cs^+ or Ba^+ incorporated stoichiometrically into the wide-bandgap lattices of LiF or KCl, substituting for alkali metal ions. These ions exhibit very strong and narrow transitions[70,71] in the near-UV: the blue part of the spectrum that has been used for laser cooling of these ions placed in ion traps. The challenge is of course replacing an ion trap with a solid lattice, and at this point, I can only speculate about feasibility of such an arrangement.

One can also, of course, consider metallic artificial structures with high effective refractive index.[1,72] This concept of "artificial dielectrics"[73] has been known since the 1940s[74,75] and has been successfully applied in, for instance, radar antennas operating in the RF and microwave ranges.[76] Simply arranging metal spheres into a regular lattice, one can obtain a very high refractive index near a localized surface plasmon polariton resonance. Alternatively, layered metal structures force the electric field inside the air gaps, which increases the effective index.[77,78] These structures work very well from RF to THz,[79] where the metal losses are small since the field does not penetrate metal. But, in the near-IR and visible ranges, the losses are enormous[13] and impossible to compensate[80] without reverting to science fiction concepts. The high loss, besides being bad by itself, also reduces the key Q parameter to about 10−20, which makes a high index unattainable.

Since, even in the absence of all scattering, metal structures with less than a 100 nm feature size retain a high loss due to Landau damping;[81] the only way to reduce metal loss is by opening wide gaps below and above the conduction band. The way to achieve it, as shown in refs 82 and 83, is either by considering artificial structures with widely separated ions[82] or with (not yet grown) a novel VdW material, such as TaS_2[83] that is, the same natural or composite materials as the ones suggested for a high index. Of course, it is entirely expected, as the presence of strong and narrow resonance portends high permittivity on the low frequency side of the resonance and negative permittivity on the high frequency side (see Figure 3c,d), which would have been very useful had it not been obscured by high absorption. This region of negative permittivity, known as a Reststrahlen band, is well-known in the IR region, where resonances are associated with optical phonons, but unfortunately, the wavelength of the optical phonon resonances does not get shorter than about 7 μm in hexagonal BN.[84]

It is tantalizing to speculate on whether negative permittivity based on electronic transitions can be observed in the region of

low loss. In ref 85, negative permittivity, $\varepsilon \sim -5$, has been measured in a dye-doped polymer around $\lambda \sim 580$ nm, but the imaginary part of $\varepsilon_i \sim 4$ was too high. Note that a very large positive $\varepsilon \sim 16$ has also been measured at a longer wavelength of $\lambda \sim 600$ nm. Therefore, the same strategy can be employed to achieve a high and negative permittivity combined with low loss, and, as shown in refs 86 and 87, for most applications, such as PIC, it is preferable to operate in a high ε polaritonic regime rather than a negative ε Reststrahlen regime.

This brings us to an interesting state of affairs regarding all types of metal-containing optics in the visible and near-IR ranges, which can be best described as "metamaterial Catch-22".[88,89] Namely, with the high losses in existing metals, such contrivances as "superlens" and "hyperlens" do not live up to their lofty promises.[9,19,90−92] If, on the other hand, a really low loss metal (or other negative ε medium) is developed, then the aforementioned concepts completely lose their appeal, as more or less conventional optics with artificially high index dielectrics can provide very high resolution and magnified images in the far field and easily outperform both super- and hyperlenses. In other words, with a reduction of metal loss, the concepts of super- and hyperlensing go straight from being unrealizable to being unneeded.

CONCLUSIONS

To conclude, this paper has been conceived as less of a review, or even a tutorial-like discourse on why the refractive indices are so low, but more as a call for action. The latter should not be interpreted as a plea for everyone in sight to immediately shift the focus of their research. As I mentioned above, independent of ever reaching the final objective, the pursuit of lofty goals provides impetus to the development of novel fabrication[23] and modeling techniques[25,93] and serves the all-important goal of assuring continuity by educating new generations of researchers.[94] Yet, it is only healthy for everyone to occasionally take a broader view outside their relatively narrow field and observe less glamorous and somewhat neglected areas hiding in plain sight and waiting to be further explored. High index materials are just one of these areas.

It is my belief that even though the basic laws of physics and the values of fundamental constants comprising the Bohr radius leave very little hope for materials with indices exceeding say $n \geq 10$ in the visible and near-IR, modern methods of growth and fabrication supported by extensive modeling may, in the near future, lead to relatively modest improvements to perhaps $n \sim 5-6$ over a relatively narrow (100 nm) band. These modest improvements will, nevertheless, have a disproportionally high impact on imaging, integrated photonics, and other walks of photonics. In this work, not considering myself a bona fide condensed matter expert by far, I nonetheless have given a few directions in which the search for these materials may proceed: d-shell transitions, high anisotropy due to VdW bonding, strong excitonic transitions, and composites with widely separated metal atoms. This list is definitely far from being complete, and I am quite sure that different, and probably better, pathways will emerge in the future. Reaching the results envisioned here would require a concerted effort of condensed matter theorists, chemists, and material engineers, not to mention photonics practitioners (I have just listed the entire scope of the *ACS Photonics* target audience). As long as this essay stimulates at least some interest in mounting this effort, its modest goal can be considered accomplished.

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