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General approach to perfect and broadband absorption in thin films

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Abstract: Absorption of light in thin films is normally probabilistic. Here, we show that a film of any lossy material, with appropriate thickness, can achieve perfect absorption at any chosen wavelength. We derive the dispersion required to maintain perfect absorption across all wavelengths, show that real materials come close and demonstrate that even non-dispersive lossy materials can typically maintain >95% absorption across a spectral octave or more. This is achieved by coating a dielectric prism, where full absorption of linearly polarized and unpolarized light can be achieved with one and two coatings, respectively. Proof-of-principle experiments with chromium- and nickel-coated glass prisms show absorption exceeding 95% throughout the 480–1700 nm spectral range. We believe that the proposed method provides opportunities from photovoltaics and light detection to reflection-prevention and stealth technologies.

1. Introduction

Absorption of light is essential for photonic technologies – regardless of whether photons are used to transfer energy, transmit information, cut materials or sense molecules, they must ultimately be absorbed. In particular, photovoltaics and detectors, with applications from astronomy and quantum technology to data storage and telecommunications, rely on high quantum efficiencies and absorption in thin films, but semiconductors with suitable electrical properties do not necessarily provide high optical absorption [1–3].

Well-known approaches to increasing absorption include the modification of the absorber surface by structuring [4,5] or application of an antireflection coating [6,7]. However, these methods are generally unsuitable for ultra-thin films and increase fabrication costs.

Absorption in ultra-thin films may be controlled through interference [8]. In the case of coherent perfect absorption (CPA) [9,10] complete absorption is achieved by the interference of counter-propagating, mutually coherent electromagnetic waves incident on opposite sides of a material with suitable optical properties. CPA has been explored in contexts ranging from microwaves to optics [11,12], and acoustics [13–15], as well as in the absorption of quantum light [16]. However, most real-world applications require broadband operation, which conventional CPA does not necessarily provide.

Several strategies for broadband perfect absorption have been proposed. For example, a broadband terahertz metasurface absorber with over 88% absorption across 4.6 octaves of the electromagnetic spectrum has been demonstrated [17]. As with other metasurface designs, however, the structure's complexity is a challenge for large-scale or low-cost implementation [18–20]. An alternative is to use an ultra-thin lossy film, where vanishingly small phase accumulation during a single pass through the film yields an optical response that is spectrally flat across a wide wavelength range. For example, unstructured ultra-thin films of chromium, MoS_2 and graphite can offer near-complete absorption across octaves of the electromagnetic spectrum but require two-sided illumination with precise beam alignment [21].

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CPA-like absorption can also be achieved with single-sided illumination, as shown in several studies [22–25]. For example, a thin film on epsilon-near-zero material coated on a prism can yield very high absorption, and layering of films with different epsilon-near-zero wavelengths can broaden the absorption band [24]. However, the inherent material requirements of such implementations limit their applicability to a narrow subset of available materials.

Here we demonstrate a simple method of obtaining complete absorption in a film made of any lossy material. Specifically, we show that a film of appropriate thickness made from any chosen lossy material can provide perfect absorption at any chosen wavelength and for unidirectional illumination. We derive the dispersion required to maintain perfect absorption across all wavelengths and demonstrate that even non-dispersive lossy materials can typically maintain >95% absorption across a spectral octave or more. Such absorption can be achieved by coating a dielectric prism with the lossy film, where full absorption of linearly polarized light from a given direction may be achieved by a single coating and full absorption of unpolarized light may be achieved by two coatings. We demonstrate the key features of such perfect and broadband absorbers in proof-of-principle experiments with glass prisms coated with chromium and nickel films of nanoscale thickness in the visible to near-infrared spectral range, observing absorption exceeding 95% over a wavelength range of 480-1700nm.

2. Design and analytical description of the perfect absorber

We consider a thin absorbing film deposited on one face of a triangular prism (Fig. 1). Light illuminates the prism at normal incidence and the absorber at an angle θ_p, which is defined by the prism's geometry and must be sufficiently large for total internal reflection, i.e. $\sin(\theta_p)\sqrt{\varepsilon_p/\varepsilon_a}>1$. We ignore reflection at the prism's glass–air input interface, which can be avoided by applying an anti-reflection coating. All materials are characterized by their permittivity ε and indicated by subscript labels for prism (p), film (f) and surrounding medium such as air (a).

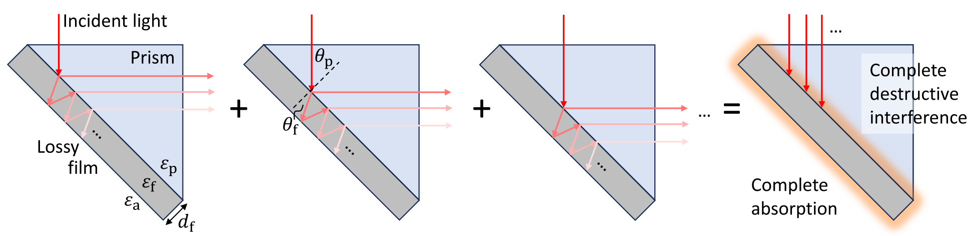

Fig. 1. Absorption mechanism. A thin film of any lossy material coated on a dielectric prism can provide complete absorption of p- or s-polarized light for appropriate choices of thickness d_f and illumination angle θ_p. Complete absorption arises from complete destructive interference. Vanishing phase accumulation with wave propagation between the interfaces of a film of deeply sub-wavelength optical thickness enables broadband absorption.

First, we derive an analytical formula for absorption of unidirectionally incident light in a lossy thin film supported by a lossless dielectric prism. Absorption occurs within the thin absorber, which can be described as a Fabry-Pérot resonator consisting of the lossy film with the prism material and air on either side of its parallel interfaces. Assuming zero material losses in the prism and a sufficiently large illumination angle to ensure total internal reflection at the absorber-air interface, absorptivity $A = 1 - R$, where R is the reflectivity. The overall reflection coefficient is given by $r_{total} = \frac{r_{pf}+r_{fa}e^{2i\delta}}{1+r_{pf}r_{fa}e^{2i\delta}}$, where r_{nm} represents Fresnel's reflection coefficient between the n and m layers, and $\delta = 2\pi\frac{d_f\sqrt{\varepsilon_f}}{\lambda}\cos(\theta_f)$ is the optical phase difference arising from a single pass through the absorbing film.

To achieve perfect absorption, r_{total} must equal 0, i.e.,

$$r_{\text{pf}} + r_{\text{fa}} e^{2i\delta} = 0 \tag{1}$$

This condition for complete absorption has a trivial solution where both terms are zero, corresponding to absence of reflection at the prism-film interface and sufficient losses within the (typically thick) film to ensure that only a negligible fraction of the incident light reaches the film-prism interface after a double-pass through the film. The physical meaning of non-trivial solutions, where both terms cancel, is that the electric field of light reflected from the prism/film interface must have the same amplitude and opposite sign compared to the electric field of the light escaping the film in order to result in complete destructive interference of all outgoing fields (Fig. 1).

Oblique incidence onto the absorbing film implies different Fresnel coefficients for *s*- and *p*-polarized light, resulting in different solutions of Eq. (1) for *s*-absorbers and *p*-absorbers, which perfectly absorb light of the respective polarization. Substituting Fresnel's formulas for r_{pf} and r_{fa} for ***s*-polarized** light, we get:

$$\frac{\cos(\theta_{\text{p}}) - A_{\text{f}}}{\cos(\theta_{\text{p}}) + A_{\text{f}}} + \frac{A_{\text{f}} - A_{\text{a}}}{A_{\text{f}} + A_{\text{a}}} e^{i\frac{d_{\text{f}}}{\lambda}4\pi\sqrt{\varepsilon_{\text{p}}}A_{\text{f}}} = 0$$

with

$$A_{\text{i}} = \sqrt{\frac{\varepsilon_{\text{i}}}{\varepsilon_{\text{p}}} - \sin^2(\theta_{\text{p}})} \tag{2}$$

Assuming that the optical thickness of the absorbing film is small compared to the wavelength and that the absorbing film's permittivity is large compared to that of the prism, Eq. (2) may be approximated (Supplement 1, Sec. 1) as

$$\varepsilon_{\text{f}} \approx \varepsilon_a + \frac{\sqrt{\varepsilon_{\text{p}}\sin^2(\theta_{\text{p}}) - \varepsilon_{\text{a}}}}{2\pi}\frac{\lambda}{d_{\text{f}}} + i\cos(\theta_{\text{p}})\frac{\sqrt{\varepsilon_p}}{2\pi}\frac{\lambda}{d_{\text{f}}} \tag{3}$$

Thus, the required relative permittivity of the s-absorber depends on the device's other material and geometrical parameters, specifically the prism angle (incidence angle onto the film) θ_{p}, film thickness d_{f}, and the prism's relative permittivity ε_{p}.

Similarly, we may expand Eq. (1) for p-polarized light:

$$\frac{\varepsilon_{\text{p}}\sqrt{\varepsilon_{\text{p}}}A_{\text{f}} - \varepsilon_{\text{f}}\cos(\theta_{\text{p}})}{\varepsilon_{\text{p}}\sqrt{\varepsilon_{\text{p}}}A_{\text{f}} + \varepsilon_{\text{f}}\cos(\theta_{\text{p}})} + \frac{\varepsilon_{\text{f}}A_{\text{a}} - \varepsilon_{\text{a}}A_{\text{f}}}{\varepsilon_{\text{f}}A_{\text{a}} + \varepsilon_{\text{a}}A_{\text{f}}} e^{i\frac{d_{\text{f}}}{\lambda}4\pi\sqrt{\varepsilon_{\text{p}}}A_{\text{f}}} = 0 \tag{4}$$

Making the same assumptions as above, this may be approximated as

$$\varepsilon_{\text{f}} \approx \frac{\varepsilon_{\text{a}}}{1 - \frac{\varepsilon_{\text{p}}}{\varepsilon_{\text{a}}}\sin(\theta_{\text{p}})} - \frac{\varepsilon_{\text{a}}}{2\pi\sqrt{\varepsilon_{\text{p}}\sin^2(\theta_{\text{p}}) - \varepsilon_{\text{a}}}}\frac{\lambda}{d_{\text{f}}} + i\frac{1}{\cos(\theta_{\text{p}})}\frac{\sqrt{\varepsilon_{\text{p}}}}{2\pi}\frac{\lambda}{d_{\text{f}}} \tag{5}$$

Thus, for the thin film to fully absorb all *s*- or *p*-polarized light, the relationship between the relative permittivities and geometry must satisfy Eq. (3) or Eq. (5). Notably, comparison of Eqs. (3) and (5) reveals that they cannot have any matching practically useful solutions, for example, the imaginary parts will only be equal for $\theta_{\text{p}} = 0$, which is incompatible with the required regime of total internal reflection. It follows – in the regime of the above approximations – that full absorption of both *s*- and *p*-polarized light cannot be achieved simultaneously with a single absorbing film. In other words, a single thin layer may perfectly absorb *s*-polarized light while partially reflecting *p*-polarized light, or vice versa. However, a pair of prisms or a prism with an *s*-absorber and a *p*-absorber on different faces can provide complete absorption of unpolarized light.

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3. Results

To verify the validity of our approximations, we compare the exact numerical solutions of Eq. (1) with the simplified forms of Eqs. (3) and (5).

Figure 2(a) shows the exact and approximate solutions for the relative permittivity of an ideal absorbing film $\varepsilon_f = \varepsilon_f' + i\varepsilon_f''$ for s-polarized light, while Fig. 2(b) shows both solutions for p-polarized light. For both polarizations, we observe good agreement between the approximate and exact solutions for thin absorbers (where $d_f \ll \lambda$), while significant deviations appear for $\lambda/d_f < 15$, when the absorbing film thickness is not much smaller than the wavelength, which is anticipated.

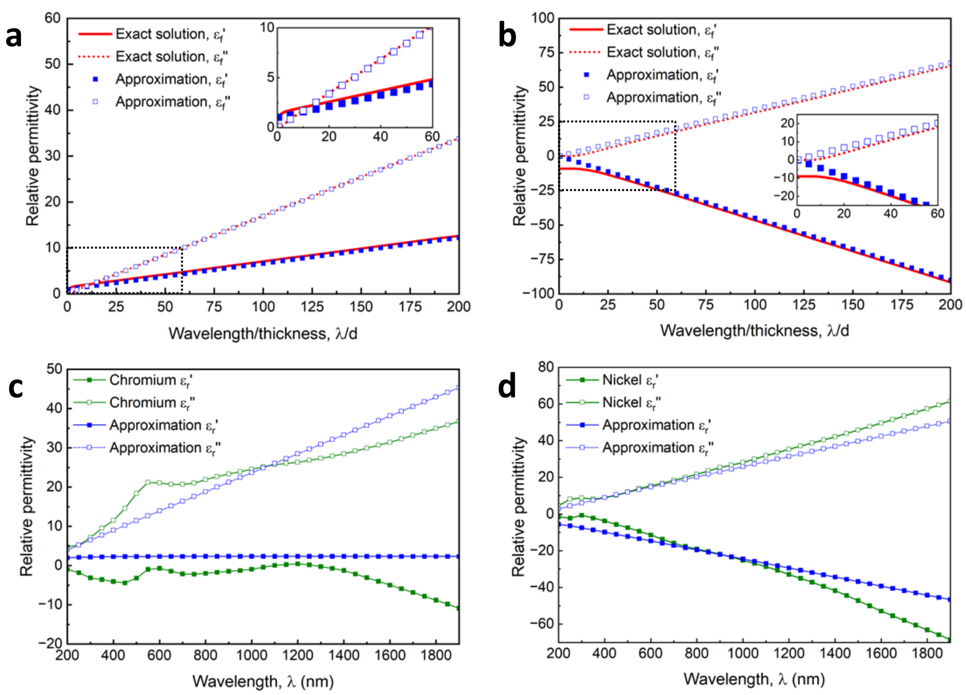

Fig. 2. Dielectric constant of ideal and real absorbing films. (a),(b) Exact numerical (red) and approximate (blue) solutions for the relative permittivity of an ideal absorbing film as a function of normalized wavelength for absorption of (a) s-polarized light according to Eq. (2) and Eq. (3) and (b) p-polarized light according to Eq. (4) and Eq. (5), respectively, assuming an angle of incidence of $\theta_p = 45°$. Insets highlight deviations that emerge when the film thickness cannot be neglected. (c) Relative permittivity of Cr compared to the ideal case for absorption of s-polarized light for 7.2 nm film thickness and $\theta_p = 42°$. (d) Relative permittivity of Ni compared to the ideal case for absorption of p-polarized light for 13 nm film thickness and $\theta_p = 48°$. All cases assume a glass prism with permittivity $\varepsilon_p = 2.25$.

According to Eqs. (3) and (5), the relative permittivity of an ideal absorbing film should be proportional to the wavelength to obtain complete absorption with unlimited bandwidth. Chromium and nickel are examples of common materials that exhibit approximately the right dispersion in the visible to near-infrared spectral range, as illustrated in Figs. 2(c) and 2(d) for s- and p-polarization, respectively. Therefore, we use chromium and nickel absorbing films for an experimental proof-of-principle demonstration of the proposed absorber.

At 1000 nm wavelength and assuming glass prisms with a refractive index of $n_p = \sqrt{\varepsilon_p} = 1.5$, we determine the film thickness and angle of incidence that maximize absorption from Eqs. (3)

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and (5) for chromium and nickel, respectively. For Cr films, these parameters are $\theta_p = 42°$ and $d_f = 7.2$ nm; for Ni, they are $\theta_p = 48°$ and $d_f = 13$ nm.

To validate the proposed concepts, we fabricated and tested devices with parameters optimized for 1000 nm wavelength as described above (Supplement 1, Sec. 2). Using a broadband light source and a spectrometer, we measured the absorption of Cr and Ni thin films over a wavelength range from 450 nm to 1700 nm (Fig. 3).

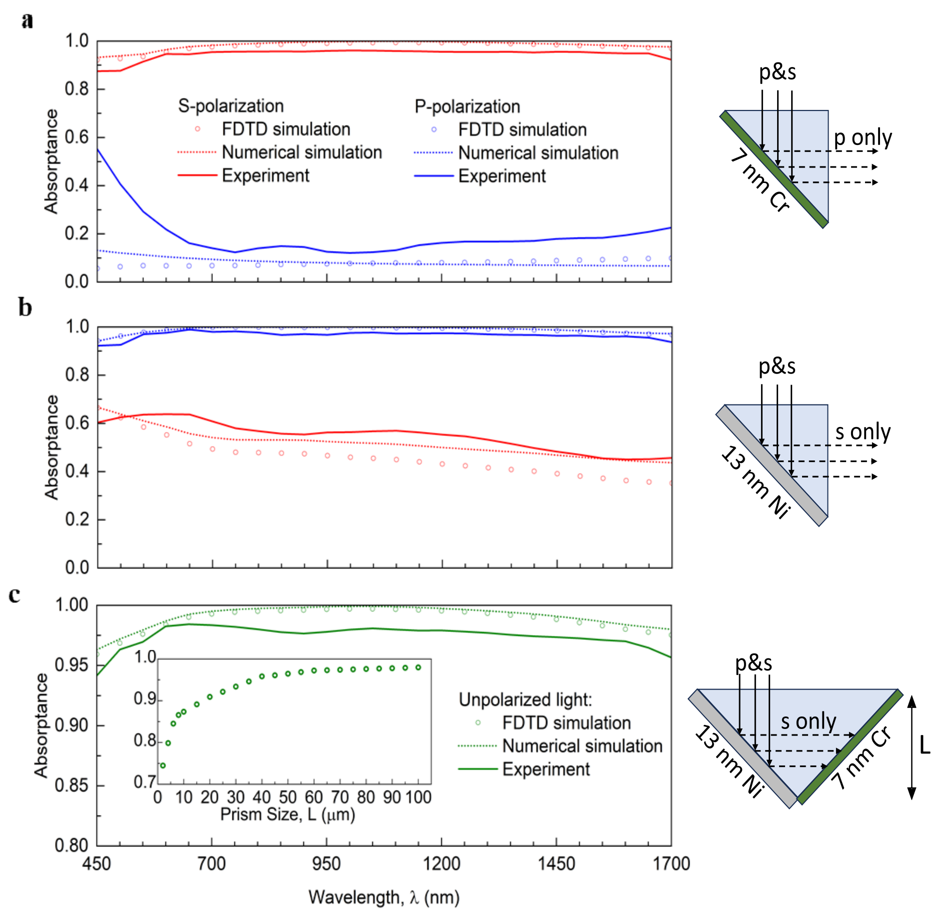

Fig. 3. Measured and simulated absorption spectra of prisms with thin-film absorbers. (a) Chromium thin film absorber device ($\theta_p = 42°$, $d_f = 7.2$ nm). (b) Nickel thin film absorber device ($\theta_p = 48°$, $d_f = 13$ nm). (c) Combined device with both absorbing thin films. (As both incidence angles add up to 90°, the films can be coated on different faces of a single right-angle prism.) The inset shows how the prism size affects absorption at 1000 nm wavelength.

According to our measurements, at 1000 nm wavelength, the Cr absorber device absorbs more than 95% of *s*-polarized light, while the Ni absorber device exhibits 96% absorption for *p*-polarized light. Theoretical results based on the transfer matrix method (TMM) and numerical finite-difference time-domain simulations (Supplement 1, Sec. 3) confirm perfect absorption for both materials at the optimization wavelength. Considering that freestanding films of 7.2 nm Cr and 13 nm Ni absorb only ~40% and ~35% of normally incident light, the enhancement in absorption is attributed to interference of light within the structure, rather than just the inherent physical properties of the absorbers.

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Importantly, absorption remains very high throughout a wide spectral range. We measured absorption exceeding 95% in the range from 700 nm to 1550 nm for the Cr film and from 550 nm to 1650 nm for the Ni film. Even though the values of relative permittivity deviate somewhat from the ideal ones according to Eqs. (3) and (5) across the measured spectral range (Fig. 2(c),(d)), the absorptivity remains high (Fig. 3). The insensitivity of absorption to changes in wavelength may be anticipated due to the thickness of the absorber being much smaller than the considered wavelengths, meaning that the wavelength-dependent phase accumulation with wave propagation through the absorber layer is negligibly small and implying that even significant changes in wavelength do not substantially affect the interference of light.

Optimal absorption of one linear polarization is accompanied by only partial absorption of the orthogonal linear polarization. For the Cr absorber device, only ~17% of *p*-polarized light is absorbed in the studied spectral range, while for the Ni absorber device ~50% of s-polarized light is absorbed. As discussed above, this discrepancy arises from different Fresnel coefficients r_{nm} for *s*- and *p*-polarized light in Eq. (1), meaning that light absorption for one polarization does not readily correlate with absorption for the other polarization. Thus, while oblique incidence onto the absorbing film introduces a degree of freedom that enables perfect absorption across a broad range of materials, it also limits optimal absorption to one eigenpolarization at a time.

As both deposited materials, Cr and Ni, serve as effective absorbers across a broad spectral range for only one polarization, the overall absorption for unpolarized light remains moderate. However, a simple combination of prisms (or both coatings on different faces of the same prism), as illustrated in Fig. 3(c), can leverage the advantages of each individual layer to provide complete absorption of unpolarized light. Consequently, the double absorber comprising both layers achieves >95% absorption across the wavelength range from 480 nm to 1700 nm in our experiments and theory and numerical modelling even predict absorption higher than 95% from 425 to at least 1925 nm. Indeed, our numerical simulations indicate that absorption exceeding 99% should be achievable across the 625–1470 nm spectral range, which spans more than one octave.

From a practical perspective, it may be desirable to fabricate arrays of small prism absorbers to obtain light-weight devices with a large aperture. Such devices could be made at low cost using imprint, angled deposition and dielectric infill techniques, however, with shrinking prism size, scattering from edges may be expected to degrade performance. Numerical simulations indicate that near-complete absorption of light is retained down to prism sizes of few tens of microns (Fig. 3(c) inset), implying that high performance can be expected from microprism arrays.

Equation (5) suggests that our design can be optimized (by choosing angle θ_{p} and thickness d_{f}) to obtain perfect absorption of at least *p*-polarized light with any lossy material and for any chosen wavelength. Figures 4 and 5 demonstrate this for *p*- and *s*-polarized light, respectively. For convenience, we assume glass prisms with a refractive index $n_{\mathrm{p}} = 1.5$ and present the optical properties of the thin film material in terms of real and imaginary parts of the refractive index, $n + ik$, where $(n + ik)^2 = \varepsilon_{\mathrm{f}}$. The highest achievable absorption (panel a) is given as a function of refractive index and may be obtained at any chosen wavelength λ for an appropriate film thickness d_{f} (panel b) and angle of incidence onto the thin film θ_{p} (panel c). To estimate the absorption bandwidth of the optimized devices, the ratio of wavelength to thickness was varied while disregarding material dispersion. The spectral range where absorption remained above 95% is shown in terms of octaves (panel d).

Importantly, complete absorption of *p*-polarized light can be obtained for all lossy materials (Fig. 4(a)). We observe three distinct absorption regimes: the regime of vanishing prism-film reflectance (red area in Fig. 4(b)), the Salisbury Screen regime (blue area in Fig. 4(d)) and the thin film regime (green area in Fig. 4(d)). These correspond to the trivial, narrow-band non-trivial and broadband non-trivial solutions of Eq. (1), respectively. The first regime takes advantage of the fact that, when the prism-film interface is fully transparent, all light at all wavelengths

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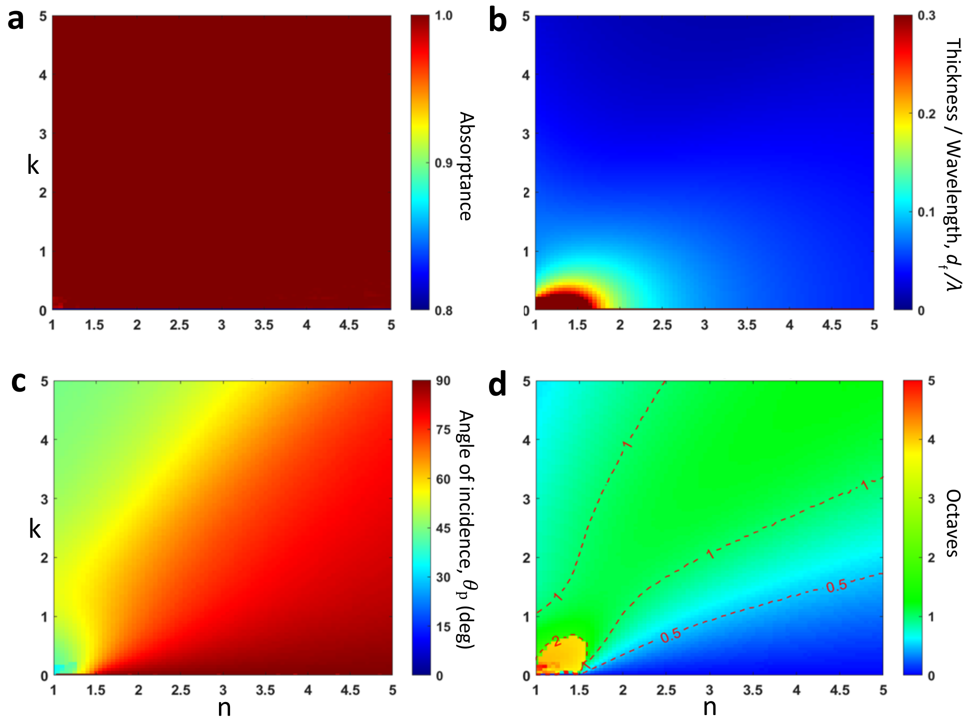

Fig. 4. Achievable absorption of p-polarized light by any lossy material. (a) Highest absorption in a film of refractive index, $n + ik$, occurring at any chosen wavelength λ for given choices of (b) film thickness d_f and (c) angle of incidence θ_p with (d) bandwidth of >95% absorption.

will be absorbed in a sufficiently thick lossy film. This situation is well-approximated for all polarizations when the refractive index of the absorber material is close to that of the prism, and for p-polarized light also at the Brewster angle. However, in practical applications, absorbers with such refractive indices are rare, and this regime requires the deposition of substantially thick films, which may not be desirable. The second regime is observed for weakly lossy materials $k \ll n$ and offers narrowband complete absorption (blue area in Fig. 4(d)) due to a non-negligible optical thickness $n\ d_f$ of the absorbing film close to $\lambda/4$ as in a Salisbury screen. Other lossy materials ($k \gtrsim n$) fall into the third regime that offers complete and broadband absorption in thin films (Fig. 4(a), (b) and (d)), which aligns well with the theory outlined above. Here, the large bandwidth arises from the small optical thickness of the film compared to the wavelength, which causes the wavelength-dependent phase accumulation with propagation through the absorbing film to be negligible, resulting in little change in interference and absorption over a large spectral range. Equations (3) and (5) are applicable in this thin-film regime.

Meanwhile, materials with parameters predicted by Eq. (3) yield perfect absorption of s-polarized light in the proposed design. As for p-polarization, also for s-polarization a regime of vanishing prism-film reflectance is observed for film materials with a refractive index that is close to that of the prism, yielding broadband and complete absorption for thick films (red area in Fig. 5(b)). Also for s-polarization, a thin film regime of perfect and broadband absorption is observed for dissimilar refractive indices of prism and film, however, in contrast to p-polarization, for s-polarization it occurs only for lossy films with $k \lesssim n$. Typically, more than one octave of absorption of more than 95% can be obtained (green area in Fig. 5(d)), but also here the

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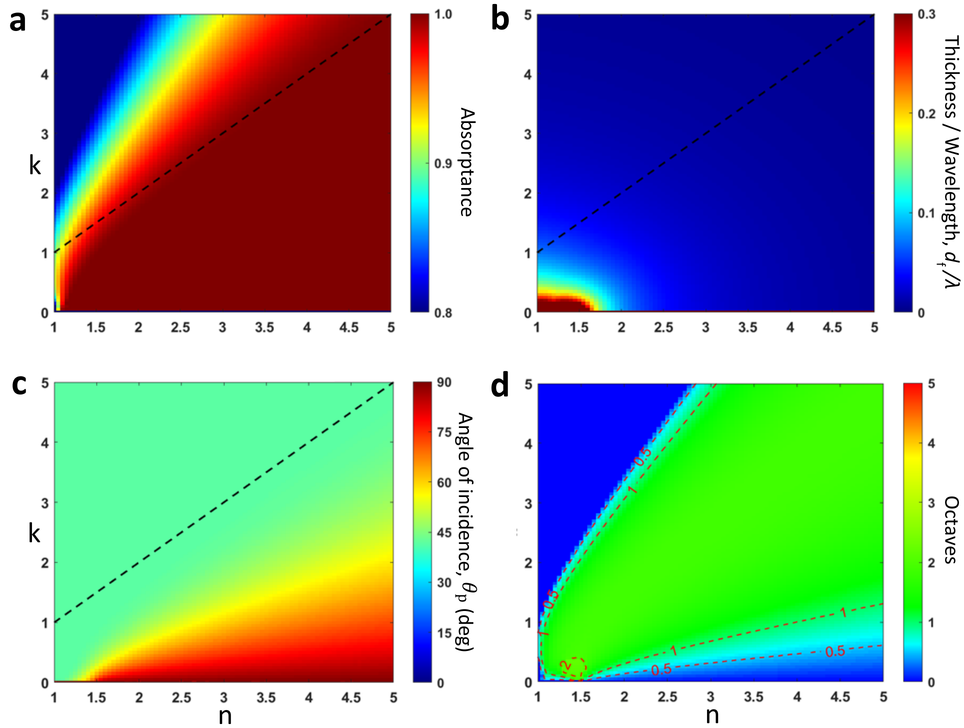

Fig. 5. Achievable absorption of *s*-polarized light by any lossy material. (a) Highest absorption in a film of refractive index, $n + ik$, occurring at any chosen wavelength λ for given choices of (b) film thickness d_f and (c) angle of incidence θ_p with (d) bandwidth of >95% absorption.

achievable bandwidth deteriorates for $k \ll n$. Film materials with $k \gg n$ do not support complete absorption of *s*-polarized light (Fig. 5(a)).

4. Summary

To summarize, we demonstrate how film thickness and illumination angle can be exploited to obtain perfect absorption of *p*-polarized light in any lossy material and perfect absorption of *s*-polarized light in any lossy material with $k \lesssim n$. We derive the dispersion required to maintain perfect absorption with unlimited bandwidth – where permittivity is proportional to wavelength – and show that even dispersionless materials can typically maintain near-perfect absorption across a spectral octave or more. Using glass prisms coated with chromium and nickel films of nanoscale thickness, we demonstrate the concept experimentally, obtaining near-complete absorption across the entire experimentally accessible 450-1700nm spectral range, which spans nearly two octaves.

Given that absorption of light is fundamentally important throughout photonic technologies and that our findings show how optimal absorption performance may be achieved with almost any material, we argue that our approach provides an opportunity to avoid common trade-offs between optical and other (e.g. electronic, thermal, mechanical) material properties. In particular, we believe that the simple yet effective approach to broadband perfect absorption reported here can be used in applications such as solar tracking photovoltaics, light sensing, reflection-prevention

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and stealth technologies. For example, the angular selectivity could be exploited to realize partially transparent solar panels that fully absorb direct sunlight.

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Disclosures. The authors declare no conflicts of interest.

Data availability. The data shown in the figures are openly available from the University of Southampton ePrints research repository [26].

Supplemental document. See Supplement 1 for supporting content.

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