

Generating Phase Singularities Using Surface Exciton Polaritons in an Organic Natural Hyperbolic Material

Philip A. Thomas¹,* William P. Wardley¹, and William L. Barnes¹

Department of Physics and Astronomy, University of Exeter, Exeter EX4 4QL, United Kingdom

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Surface polaritons (SPs) are electromagnetic waves bound to a surface through their interaction with charge carriers in the surface material. Hyperbolic SPs can be supported by optically anisotropic materials where the in-plane and out-of-plane permittivities have opposite signs. Here we report what we believe to be the first experimental study of hyperbolic surface exciton polaritons (HSEPs). We study the intensity and phase response of HSEPs in the J-aggregate TDBC (a type-II natural hyperbolic material). HSEPs can be used to generate phase singularities; the behavior of these phase singularities is a consequence of the hyperbolic nature of TDBC. The combined intensity and phase response of nonhyperbolic and hyperbolic SPs suggests that they are topologically distinct. We predict analogous effects for hyperbolic surface phonon polaritons in hexagonal boron nitride. Our Letter suggests that organic materials can provide a new platform for the exploration of hyperbolic surface polaritonics at visible frequencies.

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A key goal of nanophotonics is to rationally control the behavior of light with subwavelength structures [1,2]. One of the most widely utilized phenomena in the nanophotonic toolbox is the surface polariton (SP): the coupling of light and matter at an interface between two materials that occurs when the real components of their permittivities, $\text{Re}(\epsilon)$, have opposite signs [3]. Surface plasmon polaritons (SPPs), in which light couples to free (conduction) electrons, are the most widely studied SP and have been explored in multiple spectral bands, including the infrared [4,5], visible [3,6], and ultraviolet [7,8] regions. Many other SP modes exist [9]. Surface phonon polaritons (SPhPs), which rely on phonon modes to create spectral regions with $\text{Re}(\epsilon) < 0$, have been studied in the infrared and terahertz regions [10,11]. Surface exciton polaritons (SEPs) can occur in materials where spectral regions with $\text{Re}(\epsilon) < 0$ arise due to excitonic resonances [12,13]. Phononic and excitonic resonances can both be modeled using Lorentz oscillators [11,13], a consequence of their shared physics, in spite of their different origins.

The investigation of all-organic nanophotonics has motivated the study of SPPs in conducting polymer nanostructures, which—while currently restricted to the infrared spectral region—show promise because they can be modulated between plasmonic and dielectric behavior [14]. SEPs

provide a pathway to all-organic nanophotonics at visible frequencies since they can be generated using Frenkel excitons in organic dyes [15]. Although the fundamental physics of SEPs has been well understood for some decades [12,13], experimental work has largely been limited to the study of propagating SEP modes in planar films [15–18]. Studies of excitonic nanostructures have been predominantly theoretical [19–23] because of the challenges associated with creating organic nanostructures with sufficiently high dye concentrations to support localized SEP modes. However, a recent report of the successful creation of excitonic nanostructures shows that these challenges can be overcome and that the field might be on the cusp of a transformation [24].

Optically anisotropic materials are said to be hyperbolic if their in-plane and out-of-plane permittivities have opposite signs [25]. The unusual dispersion relations of hyperbolic materials means they can be used to design unusual optical properties such as hyperlensing [26] and negative refraction [27]. While it is possible to design artificial hyperbolic metamaterials [25], the potential of natural hyperbolic materials has also been explored, with the hyperbolic SPhPs (HSPhPs) in hexagonal boron nitride (*h*-BN) perhaps being the most well-known example [10,11]. However, very few natural hyperbolic materials have been reported at visible wavelengths [28].

In this Letter, we report what we believe to be the first experimental study of hyperbolic surface exciton polaritons (HSEPs). We show that J-aggregates can be highly anisotropic and therefore classified type-II natural hyperbolic material with giant optical anisotropy. We study prism-coupled HSEPs using spectroscopic ellipsometry, which allows us to simultaneously characterize the intensity and

*Contact author: p.thomas2@exeter.ac.uk

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phase response of HSEPs [29]. We show that HSEPs can be used to completely suppress the reflection of light, resulting in the generation of a phase singularity, and that the distinctive phase response we observe is a consequence of the hyperbolic nature of the SEPs we studied. We conduct numerical studies to consider the implications of our findings for analogous experiments using HSPHPs in the infrared. Our Letter identifies a means of studying room-temperature natural hyperbolic SPs at visible frequencies.

We studied the J-aggregate TDBC (5,6-dichloro-2-[[5,6-dichloro-1-ethyl-3-(4-sulphobutyl)-benzimidazol-2-ylidene]-propenyl]-1-ethyl-3-(4-sulphobutyl)-benzimidazolium hydroxide, sodium salt, inner salt). This dye [chemical structure in Fig. 1(a)] possesses one sharp excitonic resonance in the visible spectrum, which has made it an appealing candidate in molecular strong coupling experiments [30,31]. In high enough concentrations, TDBC films can possess a band of negative permittivity that is sufficiently negative and broad to make SEP experiments viable [15,18,24]. TDBC is part of a wider family of J-aggregates that can be used to create negative-permittivity

films at wavelengths across the visible and near-infrared spectrum [32]: while our experiments focus solely on TDBC, we expect our findings may be generalized to many J-aggregates [33]. This would allow one to use natural hyperbolic materials across a wide range of wavelengths, overcoming the main limitation of natural hyperbolic materials [34]. We discuss other candidate materials in Supplemental Material Sec. S1.1 [35].

Figure 1(b) shows photographs of one of the TDBC films made in our experiments. The optical constants of this film, determined using spectroscopic ellipsometry, are shown in Fig. 1(c) (see Supplemental Material Sec. S1.2 [35] for fabrication and characterization details). The high concentration of our film allows us to reach a minimum $\text{Re}(\epsilon) = -25$, with $\text{Re}(\epsilon) < -2$ (the condition for propagating SP modes [3]) in the spectral range 2.11–2.26 eV. These numbers are consistent with the optical constants for pure TDBC films reported by Kang *et al.* [24] and suggest that we have a broad enough spectral window to observe dispersive SEP modes in a prism coupling experiment. We studied films of thickness between 30 and 200 nm: in all cases, we found that it was only possible to achieve a good fit to ellipsometric data using an anisotropic model. J-aggregate films are structurally anisotropic, with aggregates aligning in the plane of the substrate, which should naturally give rise to optical anisotropy [36]. While our reported experiments focus on neat TDBC films, we found that this anisotropy persists for TDBC dissolved in polyvinyl acetate (see Supplemental Material Sec. S1.3 [35]). In spite of this, we are aware of only a small number of experimental works that explicitly acknowledge the optical anisotropy of TDBC [24,37], with most experiments modeling TDBC as an optically isotropic material. Additional analysis in Supplemental Material Sec. S1.4 [35] provides further support for these optical constants, suggesting that it is possible to create TDBC films with near-perfect aggregation. Since the out-of-plane permittivity is always positive, we can classify TDBC films as a type-II natural hyperbolic material in the region where the in-plane permittivity is negative (2.11–2.43 eV) [10]. Neat TDBC films also fulfil the criterion for giant optical anisotropy; see Supplemental Material Sec. S3 [35].

To characterize the HSEPs in our TDBC films, we performed a prism coupling experiment using the attenuated total reflection technique in the Kretschmann-Raether prism coupling geometry [38]. (See Supplemental Material Sec. S2.1 [35] for further details.) We characterized the HSEPs using spectroscopic ellipsometry [39], which measures ρ , the complex reflectance ratio,

$$\rho = \frac{r_p}{r_s} = \tan(\Psi)e^{i\Delta},$$

where r_p and r_s are the Fresnel reflection coefficients (amplitude) for p - and s -polarized light, respectively. $\tan(\Psi)$ is the ratio of the magnitudes of the amplitudes

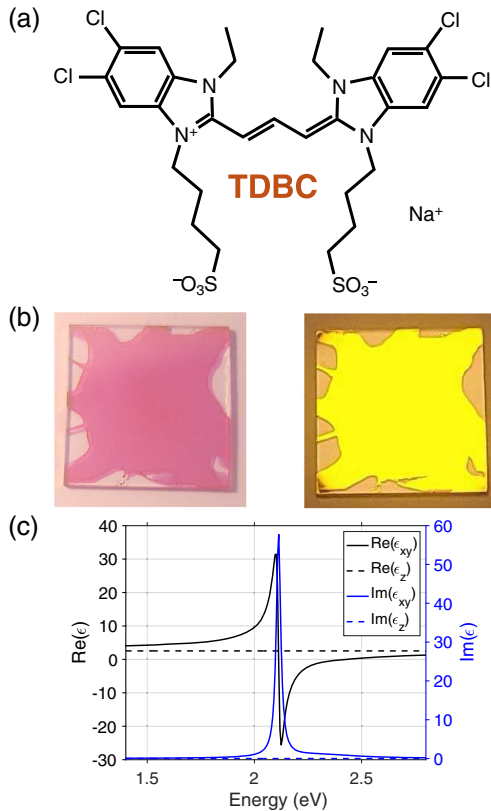


FIG. 1. (a) Chemical structure of TDBC. (b) Photograph of neat TDBC film under ambient illumination (left) and top-down illumination, reflecting light into the camera (right); the substrate is $25 \times 25 \text{ mm}^2$. (c) Optical constants of TDBC, expressed in terms of in-plane and out-of-plane permittivities (ϵ_{xy} and ϵ_z , respectively).

of p - and s -polarized light: $\Psi = 0^\circ$ when $r_p = 0$ and $\Psi = 90^\circ$ when $r_s = 0$. For propagating surface polariton modes, which are only excited by p -polarized light, we therefore expect to observe minima in Ψ . Δ is the difference in phase between p - and s -polarized light. Further details concerning ellipsometry are provided in Supplemental Material Sec. S2.1 [35].

In Fig. 2 we plot the results from our prism coupling experiments: the top row [Figs. 2(a) and 2(b)] shows results from prism coupling to a Ag film (thickness 50 nm), the middle row [Figs. 2(c) and 2(d)] shows results using a Au thin film (thickness 40 nm), and the bottom row [Figs. 2(e) and 2(f)] show results using a TDBC thin film (thickness 90 nm). Our ellipsometer has an angle range of 45° – 75° ; we have plotted our results using the incident angle of light from within the prism (45° – 65°). The left-hand column [Figs. 2(a), 2(c), and 2(e)] shows dispersion plots constructed using Ψ ; the dispersion plots in the right-hand column [Figs. 2(b), 2(d), and 2(f)] are constructed using Δ . A detailed analysis of these results, including a comparison of quality factors and an analysis of ρ plots, can be found in Supplemental Material Sec. S2 [35]. Here, we highlight three points. First, the observed SPP modes in the Ag and Au Ψ plots [Figs. 2(a) and 2(c)] closely match those

calculated using standard literature values [40]. Second, the TDBC Ψ plot [Fig. 2(e)] differs from the Ag and Au plots in two ways: there is an essentially nondispersive feature at 2.1 eV, which corresponds to the maximum in $\text{Re}(\epsilon)$ for TDBC, and the dispersive feature at 2.2–2.5 eV (replotted for this narrow spectral region in Fig. 3) is the HSEP mode. Third, we notice that all Δ plots contain phase singularities, points at which reflection is completely suppressed [41,42]. The kind of phase singularity observed here—observed not in real space, but in Fourier space—has been investigated for applications in sensing and modulation [43,44]. The Ag and Au Δ plots [Figs. 2(b) and 2(d)] both show two phase singularities generated by the SPP modes [at $(46^\circ, 2.1 \text{ eV})$ and $(58^\circ, 3.4 \text{ eV})$ for Ag and $(46^\circ, 1.8 \text{ eV})$ and $(52^\circ, 2.4 \text{ eV})$ for Au]. In contrast, there is only one phase singularity associated with the HSEP in Fig. 2(f) [see also Fig. 3(b)] [at $(53^\circ, 2.3 \text{ eV})$].

To understand the difference between SEP and HSEP behavior, in Figs. 3(a) and 3(b) we replot the Ψ and Δ dispersions in Figs. 2(e) and 2(f), rescaling to focus on the spectral region in which TDBC's HSEP mode is observed. We attempt to reproduce these plots using standard transfer matrix calculations in Figs. 3(c)–3(f). In Figs. 3(c) and 3(d), we model TDBC with the uniaxial optical constants in

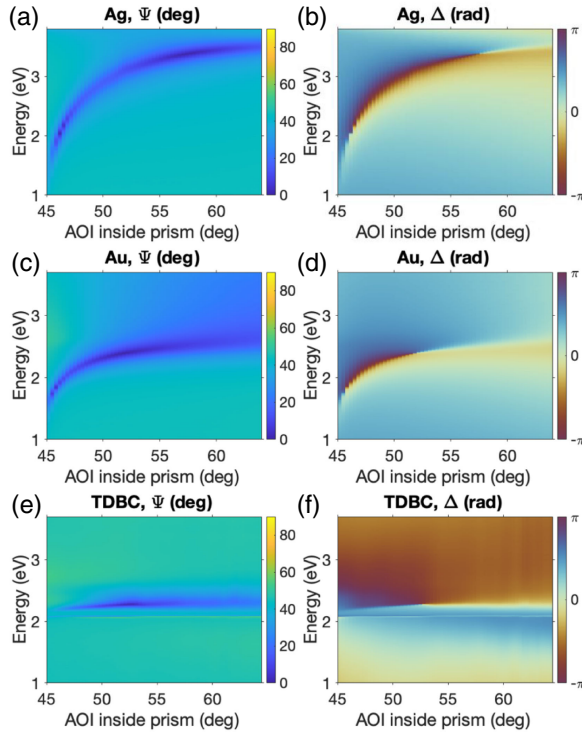


FIG. 2. Results from ellipsometric prism coupling experiments in which SPPs are observed in Ag (a),(b) and Au (c),(d) and an SEP mode is observed in TDBC (e),(f). Dispersion plots are constructed in (a),(c),(e) using Ψ (intensity) data and using Δ (phase) data in (b),(d),(f) as a function of energy and angle of incidence (AOI) inside prism. The TDBC dispersions are replotted in Fig. 3 with a rescaled energy axis.

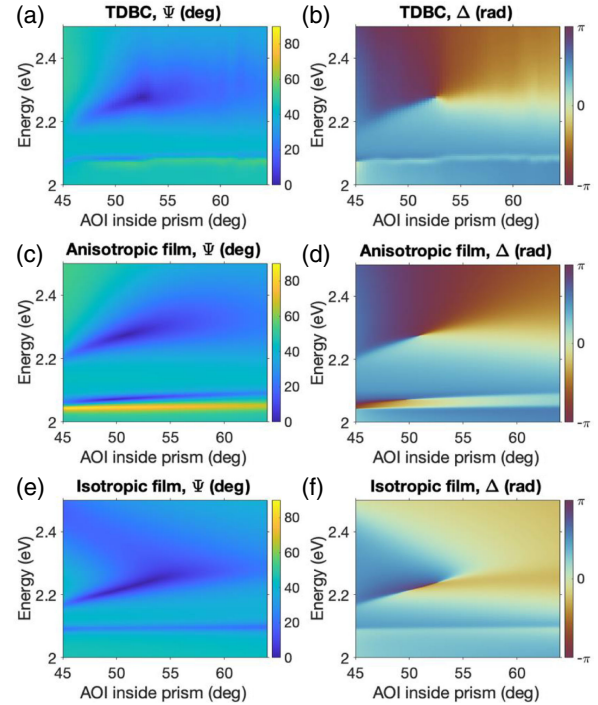


FIG. 3. The effect of anisotropy on the SEP dispersion. (a), (b) Experimental Ψ and Δ dispersions from TDBC prism coupling experiments [previously plotted in Figs. 2(e) and 2(f)]. (c),(d) Calculated Ψ and Δ dispersions using a uniaxial model of TDBC [optical constants in Fig. 1(c)], with TDBC film thickness set to 90 nm. (e),(f) Calculated Ψ and Δ dispersions using an isotropic model of TDBC [$\epsilon_{x,y}$ in Fig. 1(c)], with TDBC film thickness set to 44 nm.

Fig. 1(c) and find good agreement with experiments: the HSEP mode in Ψ and its associated phase singularity in Δ are both accurately reproduced. The two modes between 2.0 and 2.1 eV (quasinormal modes associated with the narrow region of giant optical anisotropy; see Supplemental Material Sec. S3 [35]) are also reproduced, although they appear substantially weaker in experiments (likely due to roughness of the films). In Figs. 3(e) and 3(f), we model TDBC with isotropic optical constants [using the in-plane values from Fig. 1(c)]. We can reproduce the Ψ response with an SEP by reducing the TDBC thickness from 90 to 44 nm; however, this model produces two phase singularities in Δ [at (50°, 2.22 eV) and (53°, 2.24 eV)], more closely resembling the Ag and Au SPP dispersions [Figs. 2(b) and 2(d)] than the SEP dispersion, which has just one phase singularity. Furthermore, the isotropic model predicts backbending of the SEP mode in the region 2.3–2.5 eV, which is absent in the experimental data and the uniaxial calculations. The isotropic model predicts one TM mode associated with giant optical anisotropy (a minimum in Ψ), while the experimental and uniaxial plots both show an additional TE mode (a maximum in Ψ). These calculations show that, although there are clear differences between SEPs and HSEPs in intensity measurements, the most striking differences occur in phase measurements: SEPs produce two phase singularities, while HSEPs produce only one. This suppression of reflection can be considered a form of critical coupling [45]. The conditions for zero reflection within a structure can be rationally designed by carefully tuning its material or geometric properties [46,47]. Hyperbolic materials are inherently more complex than their isotropic counterparts, meaning that the conditions for critical coupling are stricter (especially when a material has no out-of-plane absorption). Therefore, it is unsurprising that HSEPs produce fewer phase singularities than isotropic SEPs. In Supplemental Material Sec. S4 [35] we analyze ellipsometry for TDBC films with a wide range of thicknesses (30–200 nm) in the nonprism geometry (i.e., air superstrate/TDBC/glass substrate). While anisotropy radically alters the dispersion of SEP modes, these results suggest that the effect of anisotropy on other photonic modes in these structures is much more subtle.

While our experiments have focused on HSEPs, the universal nature of the Lorentz oscillator model means our findings should generalize to other hyperbolic SPs. In Fig. 4 we plot the calculated dispersion of prism-coupled HSPhPs in *h*-BN. We have performed our calculations for the spectral region for which *h*-BN is a type-II natural hyperbolic material [10]; see Supplemental Material Sec. S5 [35] for further details. In Figs. 4(a) and 4(b) we plot Ψ and Δ dispersions for uniaxial *h*-BN and observe an HSPhP dispersion in Ψ with a single associated phase singularity in Δ [at (27°, 1510 cm⁻¹)], closely resembling the experimental and calculated HSEP dispersions in

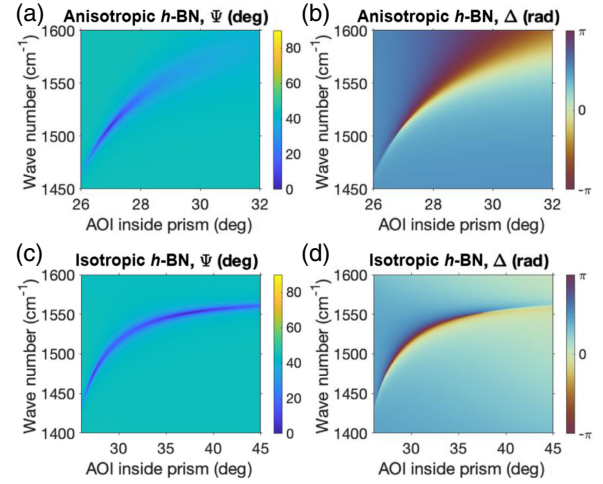


FIG. 4. The effect of anisotropy on prism coupling to HSPhPs in *h*-BN. (a),(b) Calculated Ψ and Δ dispersions using a uniaxial model of *h*-BN, with *h*-BN film thickness set to 1.5 μ m. (c), (d) Calculated Ψ and Δ dispersions using an isotropic model of *h*-BN (setting $\epsilon_z = \epsilon_{x,y}$), with *h*-BN film thickness set to 800 nm.

Figs. 3(a)–3(d). When we model *h*-BN as an optically isotropic material in Figs. 4(c) and 4(d), the nonhyperbolic SPhP dispersion much more closely resembles the Ag SPP dispersion in Figs. 2(a) and 2(b) [both in terms of the dispersion in Ψ and the observation of two phase singularities in Δ at (27°, 1470 cm⁻¹) and (38°, 1550 cm⁻¹)]. The strong parallels between our HSEP and HSPhP results suggest that HSPhPs might be used to generate phase singularities in the infrared [29]. They also suggest that HSEPs in J aggregates such as TDBC might provide a viable platform to study hyperbolic SPs at visible wavelengths.

In Fig. 5 we combine Ψ and Δ to plot the values of ρ corresponding to the hyperbolic and nonhyperbolic SP modes calculated in Figs. 3(c)–3(f) and 4 (see Supplemental Material Sec. S6 [35] for details of extraction

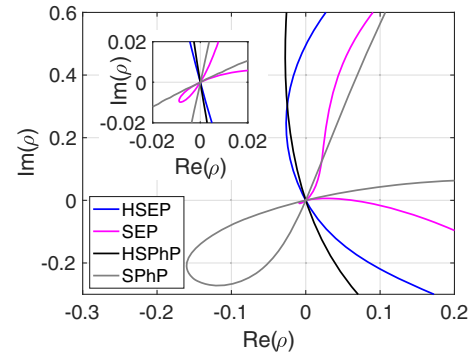


FIG. 5. The combined amplitude and phase responses of HSEP (blue line), SEP (magenta line), HSPhP (black line), and SPhP (gray line) modes. Inset: enlargement of $\rho = 0$ confirms that the plotted SEP and SPhP have the same topology.

method). The isotropic SEP and SPhP modes each generate two phase singularities, meaning that they cross $\rho = 0$ twice, creating a loop in ρ . (The SEP loop is smaller because the two phase singularities are much closer together in Fourier space, meaning the SEP amplitude stays much lower between the two $\rho = 0$ points.) Meanwhile, since the HSEP and HSPhP modes each only support one phase singularity, they only cross $\rho = 0$ once, eliminating this loop. The clear difference between the topologies of hyperbolic and nonhyperbolic SPs (a consequence of their different phase responses) perhaps hints at the more profound differences between hyperbolic and nonhyperbolic SPs [48]. While beyond the scope of this Letter, a similar approach could be used to study the response of metasurfaces, which are frequently inherently anisotropic [49].

In conclusion, we have introduced the idea of hyperbolic surface exciton polaritons and shown that they can be supported by J-aggregates, an overlooked example of a natural hyperbolic material. We have shown for the first time that surface exciton polaritons can be used to generate phase singularities; our calculations suggest that a similar phase response can be generated using surface phonon polaritons. Our combined intensity and phase analysis has revealed that hyperbolic and nonhyperbolic surface polaritons are topologically distinct. Our Letter shows that organic natural hyperbolic materials provide a viable platform for the room-temperature study of hyperbolic surface polaritons in the visible spectrum.

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Data availability—The data that support the findings of this article are openly available [50].

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