Research article

Krzysztof M. Czajkowski, Maria Bancerek, Alexander Korneluk, Dominika Świtlik and Tomasz J. Antosiewicz*

Polarization-dependent mode coupling in hyperbolic nanospheres

<https://doi.org/10.1515/nanoph-2021-0247>

Received May 18, 2021; accepted July 2, 2021;

published online July 21, 2021

Abstract: Hyperbolic materials offer much wider freedom in designing optical properties of nanostructures than ones with isotropic and elliptical dispersion, both metallic or dielectric. Here, we present a detailed theoretical and numerical study on the unique optical properties of spherical nanoantennas composed of such materials. Hyperbolic nanospheres exhibit a rich modal structure that, depending on the polarization and direction of incident light, can exhibit either a full plasmonic-like response with multiple electric resonances, a single, dominant electric dipole or one with mixed magnetic and electric modes with an atypical reversed modal order. We derive conditions for observing these resonances in the dipolar approximation and offer insight into how the modal response evolves with the size, material composition, and illumination. Specifically, the origin of the magnetic dipole mode lies in the hyperbolic dispersion and its existence is determined by two diagonal permittivity components of different sign. Our analysis shows that the origin of this unusual behavior stems from complex coupling between electric and magnetic multipoles, which leads to very strong scattering or absorbing modes. These observations assert that

hyperbolic nanoantennas offer a promising route towards novel light–matter interaction regimes.

Keywords: hyperbolic dispersion; multipole decomposition; nanoparticles; T matrix.

1 Introduction

The optical response of a particular system to external illumination is governed not only by the oscillator strength of the systems' electrons [1], but also by their spatial distribution. Indeed, structure alongside composition are the two knobs which determine a desired response or functionality. At the single-particle level, the initial choice is the use of metals or dielectrics. The surface plasmon polariton, an inherent material resonance, uses very efficiently the collective response of many conduction electrons to shape the electromagnetic field at the nanoscale, enabling, for example, plasmon-enhanced photochemistry [2], subwavelength confinement of light [3], single-molecule biosensors [4], and antennas for detection of volatile chemical species [5]. An equally rich optical response is possible with dielectric nanostructures; however, theirs is fundamentally a geometrical resonance. They simultaneously support both electric and magnetic resonances and can be chosen to have small or negligible dissipation [6]. These were subsequently observed in spherical nanoparticles [7, 8], whose response is characterized by a high degree of symmetry and allows to capture the fundamental properties of scatterers made of a particular type of material. The presence of both types of multipoles allows for significant tunability of the optical response already in simple structures [9, 10]. Some of the more spectacular effects include unidirectional scattering from nanodisks via generalized Kerker effects [11], leading to high-efficiency Huygens metasurfaces [12] or magnetic mirrors [13].

The above-mentioned examples of physical effects and/or applications are obtained by tuning the size and shape of plasmonic or dielectric nanostructures [9], all the

Krzysztof M. Czajkowski and Maria Bancerek contributed equally.

*Corresponding author: Tomasz J. Antosiewicz, Faculty of Physics, University of Warsaw, Pasteura 5, Warsaw 02-093, Poland, E-mail: tomasz.antosiewicz@fuw.edu.pl. <https://orcid.org/0000-0003-2535-4174>

Krzysztof M. Czajkowski, Maria Bancerek, Alexander Korneluk and Dominika Świtlik, Faculty of Physics, University of Warsaw, Pasteura 5, Warsaw 02-093, Poland, E-mail: krzysztof.czajkowski@fuw.edu.pl (K. M. Czajkowski), Maria.Bancerek@fuw.edu.pl (M. Bancerek), a.korneluk3@student.uw.edu.pl (A. Korneluk), dominika.switlik@gmail.com (D. Świtlik). <https://orcid.org/0000-0001-9106-2837> (K. M. Czajkowski), <https://orcid.org/0000-0001-7061-7020> (M. Bancerek), <https://orcid.org/0000-0002-7347-6320> (A. Korneluk), <https://orcid.org/0000-0002-8007-2105> (D. Świtlik)

while utilizing isotropic materials [14, 15]. In such structures, the optical response can be made strongly dependent on the direction of incident light and/or its polarization [16]; however, even more freedom in designing a desired functionality can be obtained by using anisotropic materials. At the very basic level an illumination/polarization-dependent optical response can be achieved not by tuning the shape of the particle [17, 18], but rather the birefringence of the constituent material [19, 20].

A particularly interesting class of anisotropic materials are hyperbolic ones, whose permittivity (or, in principle, also permeability) tensor is not only diagonal, but one of the principal components is opposite in sign to the other two, leading to unbounded, hyperbolic isofrequency surfaces and supporting high- k modes [21]. Hyperbolic metamaterials are presently an active research field, with investigated topics encompassing spontaneous emission [22], waveguides [23], subdiffraction focusing and imaging [24], and others. This type of dispersion can be found in naturally existing van der Waals (vdW) materials [25] such as hexagonal boron nitride (hBN) [26], WTe_2 [27], and current research is directed towards searching for new ones [28]. However, a viable alternative to obtaining hyperbolic dispersion is to use artificial metal–dielectric structures such as continuous film [29] or fishnet-type [30] multilayers or nanorod arrays [23].

The novel functionalities of large-scale hyperbolic materials [21] can be extended or adapted to nanoparticle-based optical cavities. One of these properties is confinement of optical modes, which was initially realized in stacked Ag/Ge multilayers [32], although due to large losses the quality factor of the resonances was quite low. This can be alleviated by employing low-loss materials, such as nanostructures composed of hBN, which features two reststrahlen bands with hyperbolic dispersion of different type in the mid-IR. While the low-losses of hBN are very beneficial and transient tuning of permittivity is possible [33], use of natural hyperbolic vdW materials restricts the parameter space of material properties. However, recent advances in stacking transition metal dichalcogenide (TMD) heterostructures [34] hint at the possibility of overcoming such limitations. Alternatively, at-fabrication tunability of the birefringence of hyperbolic materials is enabled by use of engineered metal/dielectric multilayers via various lithographic techniques that utilize physical vapor deposition (PVD) methods to alternately deposit both materials [35] or etch nanostructures in prepared multilayers [36]. Indeed, optical type II hyperbolic nanoantennas have been recently shown to exhibit simultaneously radiative and nonradiative modes [35] as well

as offer enhanced photoluminescence [37] and nonlinear emission [38].

The above-discussed interest in hyperbolic nanoantennas and potential use in optical devices makes it necessary to understand, at the fundamental level, the behavior of optical modes of these structures. While discussion on basic properties of anisotropic resonators can be found in the literature [39, 40] and is relevant to the topic at hand, hyperbolic nanoparticles need separate treatment. Here, we contribute to this topic by discussing basic electromagnetic properties of hyperbolic nanospheres (HNSs). The motivation behind studying a spherical object is the unambiguity of the hyperbolic origin of the optical properties, which are not masked by shape-induced anisotropy. Using a combination of finite-difference time-domain (FDTD) modelling, Mie theory, quasistatic (QS), and T -matrix analysis, we explore their modal properties and elucidate their unique spectral response.

2 Results and discussion

The hyperbolic material which comprises the studied nanoparticles is based on an artificial metal–dielectric multilayer to, on one hand, closely relate to the recently studied hyperbolic $\text{Au-SiO}_2/\text{Al}_2\text{O}_3/\text{TiO}_2$ nanostructures [35, 41] and, on the other hand, retain the ability to freely tune the birefringence by changing the composition. Thus, we assemble the hyperbolic tensor ϵ by combining a dispersionless dielectric with refractive index n_d and silver [31]. The nanoresonators, here spherical in shape, are schematically represented in Figure 1a with $\epsilon_{xx} = \epsilon_{yy} \equiv \epsilon_{\perp}$ and $\epsilon_{zz} \equiv \epsilon_{\parallel}$. Here, the subscripts $\parallel$ and $\perp$ indicate components parallel and perpendicular to the anisotropy axis, respectively, and the tensor elements are given as [42]

$$\epsilon_{\parallel} = \frac{\epsilon_m \epsilon_d}{(1 - f_m) \epsilon_m + f_m \epsilon_d}, \quad (1a)$$

$$\epsilon_{\perp} = (1 - f_m) \epsilon_d + f_m \epsilon_m, \quad (1b)$$

where f_m is the filling fraction of metal with permittivity ϵ_m and $\epsilon_d = n_d^2$.

For a metal fill factor $f_m = 0.5$ the exemplary dispersion is plotted in Figure 1b, which illustrates the presence of two types of isofrequency surfaces [21] with either two (below ~ 3.4 eV) or one (~ 3.4 – 3.8 eV) permittivity tensor elements being negative. For a larger f_m , ϵ tends to that of isotropic silver, while for smaller f_m towards an isotropic dielectric.

Previous research has proven that an effective medium description of a multilayer can be adequate for bulk

Figure 1: (a) Scheme of a uniaxial spherical nanoparticle which is composed of alternating dielectric ($n_d = 1.5$) and silver layers. In the chosen coordinate system, we then define the parallel $\epsilon_{xx} = \epsilon_{yy} \equiv \epsilon_{\perp}$ and perpendicular $\epsilon_{zz} \equiv \epsilon_{\parallel}$ permittivities, for which a metal fill factor $f_m = 0.5$ is plotted in panel (b). The permittivity of silver, based on Palik [31], is plotted for reference. For energies below ca. 3.5 eV the nanosphere is characterized by a hyperbolic dispersion relation. (c and d) Real and imaginary parts of permittivity $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ are used throughout this work as a function of f_m . The dashed lines mark 0 and split $\epsilon(\hbar\omega, f_m)$ into different dispersion regimes.

structures. The former is, naturally, a much more efficient approach to calculate optical properties; however, we need to ensure that this equivalency will remain also for small nanostructures. To that end we first use accurate FDTD simulations with mesh sizes down to 5 Å and layer thicknesses in the range 1–5 nm to confirm their convergence. For simplicity, we explicitly neglect a number of material-related effects reported in the literature, such as dependence of permittivity of thin layers on their thickness [43, 44], nonlocality [45, 46], quantum Landau damping [47], or surface roughness [48, 49]. This is motivated by our objective, namely to elucidate the dependence of optical properties of HNS on the anisotropy of the constituent material, which is assumed to be given.

The optical spectra (from FDTD) for three unique plane wave incident/polarization combinations are then plotted in Figure 2 for a sphere with radius $r = 50$ nm, $f_m = 0.5$, and 4 nm layers and compared to T -matrix calculations (open-source code SMUTHI [50–52]) for an effective hyperbolic permittivity. The agreement between the scattering and absorption spectra, Figure 2a–f, is excellent, and shows only small underestimation of some of the resonance amplitudes. This agreement makes it possible to use, throughout the rest of this work, the more efficient T -matrix method for all considered nanoparticles. Furthermore, the electric field cross sections at selected resonances in Figure 2g–i show good agreement as well, both inside and outside the HNS, especially considering the qualitatively different structures.

The benefit of the T -matrix method is that it inherently provides the response of all relevant multipoles and all

coupling elements between them. In Figure 2a–f we also plot contributions from three active multipoles, namely a strong electric dipole (ED), a weaker electric quadrupole (EQ), and the lone magnetic dipole (MD).

In Figure 2a and b the wave vector of the incident field is along the anisotropy axis, making the orientation of the linear polarization irrelevant. In the hyperbolic dispersion range the resonator has a dominant ED resonance at ~ 2.5 eV, which shows up in scattering and absorption, and a second resonance at ~ 1.7 eV, which consists of an MD and EQ. This second resonance is quite unique in that its scattering efficiency is much smaller than its absorption. This is in agreement with the experimental observation made for hyperbolic nanodisks by Maccaferri et al. [35].

The second and third illuminations conditions have the incident wave vector perpendicular to the anisotropy axis with the electric field either polarized perpendicular (Figure 2c and d) or parallel to the anisotropy axis (Figure 2e and f). In the former case only the ED and EQ multipoles are present in a spectral arrangement which is reminiscent of the response of an isotropic plasmonic particle. Also note, that the position of the ED matches that of the ED in the first case shown in Figure 2a and b. In the latter case in Figure 2e and f one sees a strong resonance ~ 3.6 eV which consists of the ED, MD, and EQ. Additionally, a second absorptive resonance (with practically no scattering) is seen at ~ 1.7 eV, which is qualitatively similar to the one at the same energy in Figure 2a and b. However, an interesting difference is seen in the sign of the contributions of the MD and EQ multipoles, which at this particular resonance are excited either in or out of phase.

Figure 2: (a–f) Comparison of scattering and absorption of a hyperbolic sphere ($r = 50$ nm): the FDTD method with explicit Ag–dielectric multilayers ($f_m = 0.5$) versus the T -matrix method for an effective permittivity. In the three unique incidence/polarization cases the two methods agree quantitatively. Here, three multipoles are active: the ED, MD, and EQ, and appear at various energies depending on illumination. For example, for $E_{||}$ a strong ED appears at 2.5 eV, but is also present above 3.5 eV and is mixed with the MD and EQ. The MD is excited only for $k_{\perp}$, $E_{||}$ and $k_{||}$, $E_{\perp}$ but is, respectively, intermixed with the EQ in- or out-of-phase. This mixing creates a unique resonance which is very strongly absorptive and has minute scattering. (g–i) Comparison of electric field cross sections from FDTD and T -matrix calculations at selected resonances further demonstrates good agreement between the two approaches, validating the use of an anisotropic effective permittivity.

2.1 Origin of dipolar modes of a hyperbolic nanosphere

While the existence of an unusual dipolar response of a type II hyperbolic nanoantenna has been reported earlier [35] and confirmed above, in this section we elucidate the origin of their nontrivial radiative and nonradiative properties. For simplicity we limit the analysis to spheres small compared to the wavelength (with $x \equiv kr \ll 1$, where k is the wavenumber), and assume vacuum as the surrounding medium. We begin by expanding the internal modes of the particles into plane waves, following Kiselev et al. [53], to obtain semi-analytical expressions for the T -matrix of the scatterer. This enables us to find the eigenfrequencies of the ED and MD resonances and discuss the scattering albedo of small hyperbolic spheres.

The boundary problem, which relates the incident and surface fields of the T -matrix of an anisotropic sphere, is formulated as Q -integrals [54]. We briefly recall the formal

notation in which each multipole is characterized by three numbers (τ, m, l) . Here τ denotes the type of the multipole with $\tau = 0$ being a magnetic and $\tau = 1$ an electric one, l is the order (1 – dipole, 2 – quadrupole, etc.) and m is the azimuthal mode number. The T -matrix for each pair of multipoles is defined as 2×2 blocks given by equation

$$T_{m_1, l_1, m_2, l_2} = -Q_{m_1, l_1, m_2, l_2}^1 \left[Q_{m_1, l_1, m_2, l_2}^3 \right]^{-1}. \quad (2)$$

The $(1, 1)$ element of the Q^3 matrix is an integral of the form

$$Q_{m_1, l_1, m_2, l_2}^{3, (1, 1)} = \frac{ix^2}{\pi} \int \left[m_r \left(\hat{\mathbf{r}} \times \mathbf{X}_{m_1, l_1}^h \right) \cdot \mathbf{M}_{-m_2, l_2}^3 + \left(\hat{\mathbf{r}} \times \mathbf{X}_{m_1, l_1}^e \right) \cdot \mathbf{N}_{-m_2, l_2}^3 \right] dS, \quad (3)$$

where $\mathbf{M}$ and $\mathbf{N}$ are the vector spherical wave functions, $\hat{\mathbf{r}}$ is the unit vector normal to the sphere surface, $\mathbf{X}^e$ and $\mathbf{X}^h$ are internal modes of the anisotropic sphere. Changing the first Q^3 matrix index leads to exchange of $\mathbf{M}$ for $\mathbf{N}$ and

vice versa. Changing the second index leads to replacing the $\mathbf{X}$ internal modes with the $\mathbf{Y}$ internal modes as shown in [54]. Expressions for Q^1 are similar, but with $(\mathbf{M}^3, \mathbf{N}^3)$ replaced by $(\mathbf{M}^1, \mathbf{N}^1)$. The internal modes of the anisotropic sphere are described in terms of plane waves, which are parametrized by angles (α, β) of the plane wave vector in spherical coordinates. The detailed expressions for those modes are presented in [54].

In general, solutions of both internal modes and Q -integrals require numerical integration and matrix inversion techniques. However, a key observation that enables us to reduce the problem complexity is that the number of plane waves required to describe the modes of an anisotropic sphere is limited. It is sufficient to replace the integral over the polar angle by only two polar angles, while the azimuthal integral is carried out analytically and reasonable approximations for small spheres can be obtained,

$$\int_0^\pi f(\beta) \sin \beta d\beta \approx \sum_i f(\beta_i) \sin \beta_i w_i. \quad (4)$$

The chosen angles are $\beta_1 = \pi/4.75$, $\beta_2 = \pi - \pi/4.75$ and the weight is equal to $w_1 = w_2 = \pi/2$ as dictated by the Gauss–Legendre quadrature algorithm. Due to symmetry only a single term of the sum has to be evaluated, while the other is identical and the summation becomes trivial. The integration over the particle surface can be performed analytically once the anisotropic sphere modes are further approximated using Taylor expansion of Bessel functions. After performing the integration, the denominator and numerator of the resulting fractions are expanded into Taylor series.

In the limit of small anisotropic nanoparticle size, the electric dipole is not coupled to any other resonance and can be considered in a purely QS manner as shown by Bohren and Huffman [55]. The resulting expressions for the electric dipole T -matrix components are close analogues of quasi-static Mie theory approximation and the in-plane and out-of-plane permittivities separate:

$$T_{1,1,1,1}^{1,1} = -\frac{2}{3}ix^3 \frac{(-1 + \epsilon_\perp)}{2 + \epsilon_\perp} \quad (5a)$$

$$T_{0,1,0,1}^{1,1} = -\frac{2}{3}ix^3 \frac{(-1 + \epsilon_\parallel)}{2 + \epsilon_\parallel} \quad (5b)$$

Consequently, the dipolar resonance occurs if the following conditions, respectively, are fulfilled

$$\epsilon_\perp = -2 \quad (6a)$$

$$\epsilon_\parallel = -2. \quad (6b)$$

Equation (6a) corresponds to the incident field with $E_\perp$ for both $k_\parallel$ and $k_\perp$, cf. Figure 2a–d and Eq. (6b) for $E_\parallel$ and $k_\perp$, cf. Figure 2e and f. As the radius increases, depolarization and radiation increase and the resonance frequency is modified in a similar manner to that of a plasmon resonance in a metallic nanoparticle.

In contrast to the ED, the MD is more complex. In particles with magnetic permeability $\mu = 1$ it can only arise as a consequence of appropriate geometrical structuring, such as large size in dielectric nanoparticles, being essentially a geometrical resonance. Thus, at first glance a QS approach is questionable. Indeed, when taken independently, the MD cannot be accounted for in a QS treatment. However, if one follows the results presented in Figure 2, namely the simultaneous presence of the MD and EQ resonances, and considers the coupled MD–EQ peak jointly, the simple treatment is successful. Indeed, Figure 3a illustrates that for the magnetic response to show up in the spectral response, at least two multipole orders ($n_{\text{rank}} = 2$) have to be considered. Due to MD–EQ coupling, the expression for the magnetic dipole in the QS approximation is

$$T_{1,1,1,1}^{0,0} = 0.725ix^5 \frac{\Psi_1(\epsilon_\perp, \epsilon_\parallel)}{(\epsilon_\perp + 1.65\epsilon_\parallel)\Psi_3(\epsilon_\perp, \epsilon_\parallel)}, \quad (7)$$

where $\Psi_1(\epsilon_\perp, \epsilon_\parallel)$ [56] and $\Psi_3(\epsilon_\perp, \epsilon_\parallel)$ [57] are third order polynomial functions of $\epsilon_\perp$ and $\epsilon_\parallel$.

Figure 3: MD and ED dipole comparison between the QS and full T -matrix approach. (a) The T -matrix extinction spectrum (black solid line) of a hyperbolic sphere with $r = 50$ nm ($k_\parallel, E_\perp$) is reproduced correctly for the ED and MD only when accounting for 4 plane waves ($n_\beta = 4$) and the first two multipole orders ($n_{\text{rank}} = 2$). (b) The QS approximation yields good scattering albedo for the ED resonance up to $r \lesssim 20$ nm. The QS scattering albedo of the MD is overestimated in a broad size range, however, (c and d) the QS-calculated MD peak position (and the ED one) agrees very well with the T -matrix-derived one.

The magnetic resonance observed in Figure 2 and described by Eq. (7) may occur, when either of the two terms of the denominator is zero. In the present case, the relevant resonance condition is then

$$\epsilon_{\perp} + 1.65\epsilon_{\parallel} = 0. \quad (8)$$

This equation, qualitatively corresponding to the Frohlich condition for a small plasmonic sphere [and here cf. Eq. (6)], defines a material-type resonance, but has an important difference. Namely, it is only fulfilled provided that the material is hyperbolic. This is markedly different for isotropic nanospheres, in which the MD is geometrical and is captured analytically only once higher order approximations of the Mie theory are used. In contrast, here it is purely a material resonance, which results from an interplay between dipolar and quadrupolar internal fields also for relatively small particles.

The approximate solutions for the ED and MD can be also used to explain the fact that the electric mode is predominantly radiative, while the magnetic mode is strongly absorbing. The scattering albedo, which is the ratio between scattering and extinction efficiencies, is given by $|T|^2/\text{Re}(T)$. For small spheres the magnetic moment is proportional to x^5 according to Eq. (7), while the electric moment is proportional to x^3 (Eq. (5)). As $x \ll 1$, the scattering albedo is damped significantly for the MD due to small $|T|^2 \propto x^{10}$. This is indeed observed when we plot the scattering albedo for hyperbolic spheres in Figure 3b.

Further confirmation that this proportionality is the main factor influencing the scattering albedo in the small particle size limit is that in this regime absorption is the dominant extinction channel, even if the material losses are vanishingly small, as shown in Figure S1. Such behavior is predicted by both QS and full T -matrix calculations. For sufficiently large size particle size scattering becomes the dominant channel when the material losses are low, at which point the scattering albedo is determined by material losses. As a consequence of the proportionality difference (with respect to x), the scattering albedo of the ED is determined mostly by material losses for much smaller particles than the MD scattering albedo.

The scattering albedo is also a good measure of the applicability of the QS approximation. As the nanoparticle size increases, various radiative effects including depolarization [58] decrease the scattering albedo, ensuring energy conservation of the optical response. When comparing the full T -matrix scattering albedo and the analogous QS method result, we conclude that the applicability range is up to about 20 nm, see Figure 3b.

Furthermore, depolarization leads to a shift of the resonance energy. For the smallest nanoparticle size ($r = 5$ nm) considered in Figure 3c and d, the T -matrix and QS approaches agree very well in terms of both MD and ED energies and extinction amplitudes proving the validity of QS method. However, when depolarization is neglected, the resonance energies are size-independent and the QS approximation cannot reproduce the size dependence of resonance energies observed in T -matrix calculations.

2.2 T -matrix symmetry for uniaxial spheres

In order to explain the dependence of the optical response on illumination, we utilize the group theory based approach presented in [59], which enables one to find the nonzero entries of the T -matrix. Then, we study the spherical wave expansion of a plane wave to derive a set of rules that rationalize the observed response for each of the three studied illumination conditions.

A uniaxial sphere belongs to the $D_{\infty h}$ point group having continuous rotation symmetry and a horizontal plane of reflection. Indeed, just based on the symmetry of the nanoparticle it is possible gain fundamental understanding of its mode structure [60, 61]. Rotation symmetry imposes that the T -matrix is diagonal with respect to m

$$T_{m_1, l_1, m_2, l_2}^{\tau_1, \tau_2} = \delta_{m_1, m_2} T_{m_1, l_1, m_2, l_2}^{\tau_1, \tau_2}. \quad (9)$$

A given T -matrix element for negative m is the same as for the positive one if the multipole is of the same type. Otherwise, the sign is switched when changing m to $-m$

$$T_{m, l_1, m, l_2}^{\tau_1, \tau_2} = (-1)^{\tau_1 + \tau_2} T_{-m, l_1, -m, l_2}^{\tau_1, \tau_2}. \quad (10)$$

For cross-coupling the change of order of interacting multipoles (e.g. ED–magnetic quadrupole (MQ) to MQ–ED) follows

$$T_{m, l_1, m, l_2}^{\tau_1, \tau_2} = (-1)^{\tau_1 + \tau_2} T_{m, l_2, m, l_1}^{\tau_2, \tau_1} = T_{-m, l_2, -m, l_1}^{\tau_2, \tau_1}. \quad (11)$$

Finally, the presence of a horizontal plane of reflection imposes

$$T_{m, l_1, m, l_2}^{\tau_1, \tau_2} = 0, \quad (12)$$

if $l_1 + l_2 + 2|m| + \tau_1 + \tau_2$ is an odd number.

An exemplary T -matrix derived using these rules is presented in Figure 4. We observe that for each block of the diagonal part of the T -matrix are $l+1$ unique elements and that each diagonal element remains unchanged upon changing the sign of azimuthal mode m . This makes the following sense: for a dipole there are parallel and perpendicular polarizabilities, for a quadrupole there are

Figure 4: Real and imaginary parts of an exemplary T -matrix of a uniaxial nanosphere (with $D_{\infty h}$ point group) with plotted MD, MQ, ED, and EQ, and projection of the three unique incident plane waves into these multipoles and their azimuthal modes.

parallel–parallel, perpendicular–perpendicular and parallel–perpendicular etc.

Simultaneously, the T -matrix has nonzero off-diagonal elements, which mirror interparticle coupling selection rules [62]. These can be summarized as follows

1. if $\tau_1 = \tau_2$: l_1 can couple to l_2 provided that $l_1 + l_2$ is even,
2. if $\tau_1 \neq \tau_2$: l_1 can couple to l_2 provided that $l_1 + l_2$ is odd.

Stated simply, coupling between electric and magnetic multipoles can happen only if one of their orders is even and the other is odd. For such orders, coupling between multipoles of the same type requires skipping every other order.

2.3 Plane wave excitation of uniaxial spheres

To elucidate the spectra of HNS it is necessary to express the incident field in terms of vector spherical wave functions to show which modes can be excited and with which phase. Extinction is calculated from a given T -matrix as $\sigma \propto \text{Re}(\mathbf{a}^* T \mathbf{a})$, where $\mathbf{a}$ is a vector of the initial field coefficients in the (τ, m, l) basis. To facilitate analysis of the mode structure, it is convenient to recast extinction of a given (τ, m, l) mode into two parts. The first is connected with *pure mode* excitation and the second results from *coupling* between modes with various τ, m and l

$$\sigma_{m,l}^{\tau} \propto \text{Re} \left(\underbrace{|\mathbf{a}_{m,l}^{\tau}|^2 T_{m,l,m,l}^{\tau,\tau}}_{\text{pure mode}} + \mathbf{a}_{m,l}^{\tau} * \sum_{m',l',\tau'} \underbrace{T_{m,l,m',l'}^{\tau,\tau'}}_{\text{coupling}} \mathbf{a}_{m',l'}^{\tau'} \right). \quad (13)$$

The above equation underlines the significant influence of coupling on the extinction spectrum. It is determined not only by off-diagonal elements of the T -matrix,

but also the incident field coefficients, i.e. $[\mathbf{a}_{m,l}^{\tau}]^* \mathbf{a}_{m',l'}^{\tau'}$ product.

Now we focus on plane wave illumination with initial field coefficients $\mathbf{a}_{p,m,l}^{\tau}$, where we add the index p to denote the wave's polarization. For normal incidence along the optical axis, $\text{TE}_{\parallel}$ with $k_{\parallel}$ (equivalent to $\text{TM}_{\parallel}$ except for a multiplicative constant i ; both cases of normally incident light will be referred to as TEM where appropriate) the initial field coefficients are given by [54]

$$\mathbf{a}_{\text{TE}_{\parallel},\pm 1,l}^0 = -i^l \sqrt{2l+1} \quad (14)$$

$$\mathbf{a}_{\text{TE}_{\parallel},\pm 1,l}^1 = -i^{l+1} \sqrt{2l+1} (\mp i) \quad (15)$$

and zero otherwise. When the wave vector of the incident plane wave is perpendicular to the optical axis, there are two cases for the $\text{TM}_{\perp}$ and $\text{TE}_{\perp}$ polarizations. In the first case for TM with $k_{\perp}$ and $E_{\parallel}$ we have

$$\mathbf{a}_{\text{TM}_{\perp},m,l}^0 = \begin{cases} 0, & l + |m| \text{ even} \\ i \frac{(l+|m|)!! \cdot (l-|m|+1)}{(l+1-|m|)!!} \tilde{c}_{ml}, & l + |m| \text{ odd} \end{cases} \quad (16)$$

$$\mathbf{a}_{\text{TM}_{\perp},m,l}^1 = \begin{cases} m \frac{(l+|m|-1)!!}{(l-|m|)!!} \tilde{c}_{ml}, & l + |m| \text{ even} \\ 0, & l + |m| \text{ odd} \end{cases} \quad (17)$$

For the TE polarization ($k_{\perp}, E_{\perp}$)

$$\mathbf{a}_{\text{TE}_{\perp},m,l}^0 = \begin{cases} -im \frac{(l+|m|-1)!!}{(l-|m|)!!} \tilde{c}_{ml}, & l + |m| \text{ even} \\ 0, & l + |m| \text{ odd} \end{cases} \quad (18)$$

$$\mathbf{a}_{\text{TE}_{\perp},m,l}^1 = \begin{cases} 0, & l + |m| \text{ even} \\ \frac{(l+|m|)!! \cdot (l-|m|+1)}{(l+1-|m|)!!} \tilde{c}_{ml}, & l + |m| \text{ odd} \end{cases} \quad (19)$$

The above used $\tilde{c}_{ml}$ constant is derived and defined in the Supporting Information (SI). Using the above coefficients we can discuss how the excited modes depend on the incident field polarization and wave vector direction.

Regardless of polarization and wave vector, both electric and magnetic multipoles are always excited and all orders enter the expansion, while the azimuthal modes are governed by the zeros of the Legendre polynomials and their derivatives are shown in the SI. For normal propagation, $k_{\parallel}$, only modes with $|m| = 1$ can be excited. Alternatively, for $k_{\perp}$ the selection rules can be deduced as follows. First, assuming $\text{TE}_{\perp}$ polarization:

1. If l is odd, even azimuthal modes are excited for magnetic modes and odd ones for electric modes;
2. If l is even, odd azimuthal modes are excited for magnetic modes and even ones for electric modes.

Figure 5: Spectra of total extinction efficiency (solid black line) and their decomposition into five moments MD, MQ, ED, EQ, and EO vs illumination orientation and hyperbolic ($f_m = 0.5$) sphere radius. (a) For $k_{\parallel}$ (TEM polarization) for small r the ED is dominant, but with increasing r a very sharp MD appears which is coupled with the EQ. However, higher electric multipoles remain weak even for $r = 120$ nm, while an additional ED appears below the MD. (b) For $k_{\perp}$ and $E_{\perp}$ ($TE_{\perp}$ polarization) the spectra evolve with r as those of a pure metallic nanosphere with first a dominant ED, followed by a stronger EQ for $r = 90$ nm, and then an EO. The MD, which is present in case (a) is absent here. (c) For $k_{\perp}$ and $E_{\parallel}$ ($TM_{\perp}$ polarization) the spectrum is of smaller amplitude and qualitatively different. The MD peak is at the same spectral location as for case (a), but couples out-of-phase to the EQ. The electric multipoles below 3 eV grow gradually with r . Interestingly, the peak above 3 eV is composed of a mix of all multipoles with higher orders appearing with increasing r . (d) Induced electric multipoles for TEM and $TE_{\perp}$ polarizations exhibit different symmetry in relation to the anisotropy axis, illustrating the same response of the ED mode and different response of the EQ mode.

For $TM_{\perp}$ polarization the opposite parity holds. The phase of the incident field for various polarizations is described by the relations

$$a_{TE}^0 = -ia_{TM}^1 \quad \text{and} \quad a_{TE}^1 = -ia_{TM}^0. \quad (20)$$

Additionally, for normal incidence $|a_{TE_{\parallel}}| = |a_{TM_{\parallel}}|$, because of the relation $a_{P_{\parallel},m,l}^0 = \text{sgn}(m)a_{P_{\parallel},m,l}^1$. The phase for the three unique polarizations is shown in Figure 4. Moreover, only an incident field with $k_{\perp}$ ($TE_{\perp}/TM_{\perp}$) with an azimuthal mode $m = \pm 1$ can be expressed in relation to normal incidence ($k_{\parallel}$) as

$$a_{P_{\perp},\pm 1,l}^{\tau} = (-1)^{l-1} a_{P_{\parallel},\pm 1,l}^{\tau}. \quad (21)$$

In practice, one can express $TE_{\perp}$ -polarized electric dipole and magnetic quadrupole as well as $TM_{\perp}$ -polarized magnetic dipole and electric quadrupole incident field

coefficients in terms of normal incidence incident field coefficients.

We shall now discuss the extinction spectra of hyperbolic nanospheres using the above relations while simultaneously investigating how they evolve with radius. We plot extinction spectra for increasing r from 30 to 120 nm in Figure 5 and in the SI for the three unique incidence/polarization cases.

For polarization $E_{\perp}$ and irrespective of $\mathbf{k}$ direction, a dominant ED is always present and of the same amplitude at a given r , cf. Figure 5a and b, as would be expected for an isotropic plasmonic nanosphere. This can be rationalized by the fact that for $E_{\perp}$ polarization only $m = \pm 1$ modes are excited [Eqs. (15) and (19)] and ED is negligibly coupled to other resonances. However, the higher electric multipoles are only visible in the plots in Figure 5b with $k_{\perp}$, while being much weaker for $k_{\parallel}$. Focusing on the EQ around 2.5 eV as an example, its amplitude for $TE_{\perp}$ polarization at $r = 60$ nm

is larger than for normal incidence for $r = 120$ nm. In fact, the spectral response of the HNS under $TE_{\perp}$ illumination shows a plasmonic-like response, while not so for TEM illumination. This qualitative difference between the two cases can be understood as follows.

The electric dipole consists of positive and negative charges induced at opposite ends of the nanosphere, as schematically illustrated in Figure 5d. If one neglects retardation and simplifies the ED to a positive and negative induced point charge, then the induced ED due to symmetry for TEM and $TM_{\perp}$ polarizations in relation to the anisotropy is identical. From an energy point of view, the *self-energy* of the two EDs is the same. However, for the quadrupoles the circumstances are different. A physical EQ consists of four point charges which are located in the plane of incidence as marked schematically in Figure 5d. For TEM incidence the EQ is arranged along the anisotropy axis, while for $TE_{\perp}$ it is perpendicular to it. Hence, due to the different symmetries of these two cases, the origin of the qualitative difference between the two EQ is clear.

The other significant difference between the TEM and $TE_{\perp}$ cases is the presence of a strong MD for $k_{\parallel}$ (Figure 5a), which is weakly radiative for small r as discussed above. Neither the MD nor other magnetic modes are present in Figure 5b ($TE_{\perp}$), when the spectra of an HNS are reminiscent of an isotropic plasmonic nanosphere for all radii (and $\lesssim 3.5$ eV). Magnetic modes are, however, present in Figure 5c ($TM_{\perp}$), specifically the magnetic one is found at the same spectral location as in Figure 5a. However, its amplitude is much weaker due to destructive interference from the coupled EQ. Conversely, in Figure 5a the MD and EQ interfere constructively (cf. Figure S3).

The origin of this different interaction between the MD and EQ modes for TEM ($k_{\parallel}$) and $TM_{\perp}$ polarizations ($k_{\perp}$, $E_{\parallel}$) is deduced from the T -matrix and the incident field, which are depicted in Figure 4. In both cases, the incident field excites the same azimuthal $m = \pm 1$ modes of both the MD and EQ, which would result in the same extinction in the absence of MD–EQ coupling. However, MD–EQ coupling contributes to extinction with different sign for $k_{\parallel}$ and $k_{\perp}$ TE incidence as predicted by Eq. (21). This same Eq. (21) also explains the switch between constructive/destructive ED–MQ coupling illustrated in Figure S3, clearly visible for $r \gtrsim 80$ nm. Detailed derivation of the sign of the electric or magnetic quadrupoles' contribution is presented in Section S2 of the SI. Hence, based on the spectra in Figures 5 and S3, it is clear, that the multipolar properties of even the most simple of hyperbolic nanoparticles, i.e. a sphere, are quite complex. In particular, due to cross coupling of electric and magnetic modes, higher order modes

can appear at energies below lower order modes of the same type.

The negative contribution of the EQ to extinction and absorption deserves a qualitative discussion. For any system composed of coupled elements its total extinction is positive. However, due to coupling (multiple scattering) within the system some resonators may receive more energy than is directly provided to them by the source. If the coupling is large enough and specifically out-of-phase with the incident field, these particles' extinction may turn negative, implying that they effectively return more energy to the electromagnetic field than they receive directly from the source [63]. While this phenomenon elucidates negative extinction in an element, it does not apply to absorption in individual (even coupled) resonators, in which it has to be positive, in particular in an HNS (cf. Figure S3). However, when the optical cross sections are decomposed into multipolar components, scattering of every multipole is always positive [54]. On the other hand, extinction of individual multipoles does not have to be larger than scattering of the same multipoles or even be positive, since they are expressed by the expansion coefficients of both the incident and scattered fields [54]. Thus, in a resonator without spherical symmetry coupling between multipoles may beget very efficient energy transfer between them, what from the outside may look like a particular mode is “generating” energy. This phenomenon, like negative extinction [63], is connected with out-of-phase coupling between the interacting multipoles, as derived in Section S2 of the SI.

2.4 Dependence on material properties

Up to this point the hyperbolic material was composed of equal amounts of metal and dielectric with $f_m = 0.5$. However, as shown in Figure 1c and d, varying f_m allows for significant tunability from a uniaxial dielectric through a type I or II hyperbolic material to a uniaxial metal. In Figure 6 we present how the extinction spectrum and its multipole decomposition evolve with f_m for an HNS with $r = 80$ nm.

It is clearly seen, that overall the dominant response is of the electric type, with the ED and MD exhibiting a strong response, especially for $TE_{\perp}$ polarization in Figure 6b, which has the typical response of a plasmonic sphere. This can be confirmed by plotting the spectra vs $\sqrt{f_m}$, which show an almost linear dependence consistent with the localized surface plasmon frequency, as well as the EQ and EO, being proportional to the carrier concentration. Also note, that while initially absent for small f_m , the EQ

Figure 6: Optical properties of hyperbolic nanospheres, $r = 80$ nm, for the three unique illumination conditions vs energy and metal filling fraction: Extinction efficiency, Q_{ext} , its decomposition into the first six moments: MD, MQ, MO, ED, EQ, and EO. Black lines mark regions with positive/negative permittivity tensor elements. Note the monotonic behavior of the mainly uncoupled electric modes with f_m , while the magnetic resonances in the hyperbolic dispersion range in all cases coupled to other modes. Furthermore, often the coupled modes of high order occur below the lower order modes of the same type, e.g. EQ below ED for TEM polarization.

and EO resonances appear for TEM and $\text{TM}_\perp$ polarizations when the material becomes a uniaxial metal.

The MD response is, however, quite strong and clearly seen in Figure 6a and c. Furthermore, in contrast to the monotonic dependence of the main electric resonances, the MD and higher magnetic order modes show a peculiar crescent-like shape. Indeed, the electric modes directly coupled to these crescent-shaped magnetic resonances show identical behavior, while simultaneously exhibiting constructive/destructive interference depending on the polarization of incident light. The magnitudes of these coupling relations are plotted in Figure 7.

A peculiar feature of the coupled electric and magnetic modes is the symmetric crescent-like profile with respect to $f_m = 0.5$. This dependence of the modes on f_m can be rationalized using our analytical model using the electric and magnetic dipoles as examples, while for higher orders the behavior will be qualitatively similar. To facilitate the analytical analysis we substitute the experimental dispersion of silver with a Drude permittivity fitted to that of silver $\epsilon_m(\omega) = \epsilon_\infty - \omega_p^2(\omega^2 + i\gamma\omega)^{-1}$ with parameters $\epsilon_\infty = 4.18$, $\omega_p = 8.76$ eV, $\gamma = 91$ meV. The material data is then substituted into Eq. (8). Assuming negligible influence of material losses on the resonance condition, we solve for the zeroing of the real part of Eq. (8) to find the resonance condition of the magnetic dipole

$$\omega_{\text{res}}^{\text{MD}} \approx \frac{\omega_p \sqrt{F_m}}{\sqrt{\epsilon_\infty F_m - \epsilon_d(1.3 + F_m + 1.6\sqrt{0.7 + F_m})}}, \quad (22)$$

where $F_m = f_m(1 - f_m)$ is a symmetric function with respect to $f_m = 0.5$. The above result is thus consistent with the

Figure 7: T-matrix derived coupling between multipoles in an anisotropic nanosphere with $r = 80$ nm and $n_d = 1.5$ whose extinction is plotted in Figure 6. The actual coupling between multipoles in Figure 6 depends on the incident field direction and polarization.

symmetric, crescent-shaped of the MD resonance, confirms the previous result as well as offers further proof of the validity of QS approximation.

An analogous approach yields the resonant frequencies of the electric dipole for perpendicular

$$\omega_{\text{res}} = \frac{\omega_p \sqrt{f_m}}{\sqrt{2 - \epsilon_d(-1 + f_m) + \epsilon_\infty f_m}} \quad (23)$$

and parallel

$$\omega_{\text{res}} = \frac{\omega_p \sqrt{2 + \epsilon_d - 2f_m}}{\sqrt{\epsilon_\infty(2 + \epsilon_d - 2f_m) + 2\epsilon_d f_m}} \quad (24)$$

polarizations. Both approach the well-established result for a quasi-static sphere $\omega_{\text{res}} = \omega_p / \sqrt{2 + \epsilon_\infty}$ in the limit of purely metallic particles and confirm the earlier observation on the quasilinear dependence of the resonance positions of the ED on $\sqrt{f_m}$. Also, for both the ED and MD modes, the eigenfrequencies are proportional to ω_p of the metal, which indicates that changing the free charge concentration of the conducting material is a direct method of tuning the properties of hyperbolic nanoparticles.

2.5 General considerations on hyperbolic nanoparticles and their permittivities

Having obtained the main goal of elucidating the optical properties of HNS, we briefly discuss a few aspects of hyperbolic materials which could be the basis for realizing hyperbolic nanoresonators. The degree of anisotropy in known natural materials was typically small until the discovery of vdW materials, while natural hyperbolicity was not common. However, with the rise of extensive research on vdW materials, hyperbolic dispersion can easily be found in the literature. Examples include hBN with two reststrahlen bands [26] or materials with Drude-like dispersion for the in-plane components, such as considered here, TaS₂ or TaSe₂ [64, 65]. While the permittivity of a vdW material depends on the number of layers, once the thickness exceeds a few tens of layers the bulk properties are established and the below-discussed challenges are alleviated. However, to gain independence from chemistry and arbitrarily (though within physical limitations) shape the hyperbolicity, the method of choice is to use structured materials in the form of metal–dielectric metamaterials formed into multilayers or wire-media [21]. Sizes of these layers (or wires) have to be small enough so that an effective medium approximation will hold; however, using thin layers can bring about certain changes. These encompass issues related to fabrication, fundamental physical effects, or both.

One important aspect is that fabrication of thin, continuous layers is challenging, especially for metals. One of the best plasmonic metals, Ag, is known for its island growth and wetting layers are needed to obtain smooth, continuous metal layers, with germanium being a prime choice [44]. This enables deposition of sub-10 nm layers with good qualities and, if needed, Ge can be used as the dielectric multilayer pair to Ag to form low-loss hyperbolic nanostructures with resonances below ~ 1 eV. As we show in Figure S4, already 10 nm layers are adequate to obtain well-formed both ED and MD–EQ resonances. Directly connected with deposition of thin layers is a question of the achievable surface roughness [48, 49], which can quickly destroy the hyperbolic-material-dependent MD–EQ resonance. Already a root mean square (RMS) of 0.4 nm for 4 nm Ag layers is enough to smear the MD–EQ (Figure S5). However, by controlling the temperature during metal evaporation an RMS of 0.2 nm for a 10 nm Ag film is obtainable, and is in fact limited only by the surface roughness of the substrate [66]. Such small RMS values are enough to retain all the important spectral features of our hyperbolic nanoresonators. Moreover, for thicker layers the impact of RMS is lower and for 10 nm Ag layers with an RMS of 0.6 nm the MD–EQ peak is easily seen (Figure S6). This means that widely used PVD and atomic layer deposition (ALD) methods are viable fabrication paths. PVD can be used in a standard lithography process to deposit the layered nanoparticle through a mask [35], while both PVD and ALD enable deposition of wafer-scale multilayers for subsequent sacrificial etching of nanostructures [36, 67]. Such a lithographic approach will yield structures with flat top and bottom surfaces with the simplest resonator being a disk. However, the optical spectra of a hyperbolic nanosphere and nanodisk are qualitatively similar (Figure S7). These spectra prove that the general behavior of hyperbolic resonators is well described by Eqs. (6) and (8), but the shape dependence is also a key parameter in determining the actual response. Finally, it is necessary to account for the fact that the permittivities of many materials deposited as thin layers differ from their bulk values [43, 44, 68].

Nonlocality, that is the dependence of material properties on the wave vector, can modify the effective medium permittivity depending on the angle of incidence [46]. In the region of epsilon near zero, additional light waves [45] or complex eigenmodes [69] can be observed, which are neglected by a local effective medium theory. However, the optical modes reported herein are observed far from the epsilon near zero range and we expect that they will not be affected. Other potentially relevant effects are tied

to the characteristic sizes of metal grains or layer thicknesses, when the movement of charge carriers is inhibited or altered. This phenomenon is observed for small dimensions of metals which modify the movement of free electrons such as size quantization or surface screening. These lead to spectral shifts and broadening of the surface plasmon [70, 71] or quantum Landau damping in thin metal layers [47].

Despite the many causes of why engineered metal–dielectric multilayers may behave differently than when modeled using a local approach, the main observations on the origin of the various optical resonances will hold. They may, of course, occur at shifted frequencies due to different hyperbolic permittivity after accounting for the various above mentioned effects. However, such changes are predictable beforehand and many of them can be circumvented by using vdW materials which are anisotropic or even hyperbolic and can easily be tuned after fabrication by electrostatic gating [72].

3 Conclusions and outlook

In this work we have presented a detailed analytical and numerical study of the optical properties of hyperbolic nanospherical antennas using an artificial silver–dielectric effective multilayer as the exemplary material. It is clear that hyperbolic dispersion enables a rich modal structure which is strongly dependent on the polarization and direction of incident light. For $TE_{\perp}$ illumination the response mirrors that of a plasmonic nanosphere with a scaled plasma frequency that is determined by the density of charge carriers, exhibiting the full spectrum of multiple electric multipoles up to the number determined by the nanosphere diameter. However, for TEM incidence only the electric dipole remains of plasmonic behavior, while below the ED a very strongly absorptive magnetic dipole is present. The MD is also present for the $TM_{\perp}$ polarization, although its optical cross section is much weaker than for the TEM case.

These modal properties are a consequence of the interplay of the T -matrix derived coupling conditions between various multipole orders, whose efficiencies are determined by the hyperbolic dispersion. The unique coupling, which is absent in isotropic nanospheres, begets the appearance of an atypical modal order. For example, very sharp electric quadrupoles occur at lower energies than the first electric dipole resonance. Similar relations are present for the magnetic response.

Furthermore, by employing a quasistatic analysis of the T -matrix of hyperbolic nanospheres we are able to

elucidate the origin of the electric and magnetic dipolar modes. With this approach we derive material-dependent resonance conditions for the ED and MD in Eqs. (6) and (8), respectively. Specifically, we prove that the unique MD mode present in the hyperbolic nanospheres is a material resonance determined by the ordinary and extraordinary permittivities and requires the sign of these two values to be opposite. It is expected, especially for the plasmon-like ED, that the conditions expressed by Eqs. (6) and (8) will need to be amended to account for nonspherical resonators by using a shape factor L . The QS approximation is also crucial in explaining recent observations [35] of why the magnetic dipole is very strongly absorptive, while in contrast the electric one radiates much more efficiently. Our analysis shows that the origin of this unusual behavior of the MD stems from complex coupling between electric and magnetic multipoles, which leads to very strongly scattering or absorbing modes depending on antenna size and dissipative losses.

Finally, we show how the optical response of hyperbolic nanoparticles can be tuned by varying the charge carrier concentration which sets the magnitude of the metallic permittivity tensor. One interesting example is the ability to tune the spectral separation between the electric and magnetic dipoles by varying the plasma frequency. The tunability of hyperbolic nanostructures' spectral response has been studied recently, e.g. in terms of anomalous scattering leading to electromagnetic cloaking [73]. Hyperbolic nanocavities are of interest in the field of strong light–matter interaction; they have been demonstrated to enhance far-field radiation and shorten lifetime of coupled quantum emitters [74]. Hyperbolic dispersion was also shown to increase sensitivity of refractometric sensors due to excitation of high- k modes [75], making such materials viable candidates for refractometric sensing purposes. Thus, we are convinced that our study offers critical insight into the electromagnetic properties of hyperbolic nanoparticles, which seem to be extremely promising candidates for novel devices enabling efficient light–matter interaction.

Acknowledgments: The computations were enabled by resources provided by the Interdisciplinary Center for Mathematical and Computational Modelling via the project #G55-6.

Author contribution: All the authors have accepted responsibility for the entire content of this submitted manuscript and approved submission.

Research funding: We acknowledge support by the Polish National Science Center via the projects 2017/

25/B/ST3/00744 (K. M. C., D. Ś., T. J. A.) and 2019/34/E/ST3/00359 (M. B.).

Conflict of interest statement: The authors declare no conflicts of interest regarding this article.

References

- [1] Z.-J. Yang, T. J. Antosiewicz, R. Verre, F. Javier García de Abajo, S. Peter Apell, and M. Käll, “Ultimate limit of light extinction by nanophotonic structures,” *Nano Lett.*, vol. 15, pp. 7633–7638, 2015.
- [2] B. Iandolo, T. J. Antosiewicz, A. Hellman, and I. Zorić, “On the mechanism for nanoplasmonic enhancement of photon to electron conversion in nanoparticle sensitized hematite films,” *Phys. Chem. Chem. Phys.*, vol. 15, pp. 4947–4954, 2013.
- [3] R. Chikkaraddy, B. d. Nijs, F. Benz, et al., “Single-molecule strong coupling at room temperature in plasmonic nanocavities,” *Nature*, vol. 535, pp. 127–130, 2016.
- [4] S. S. Aćimović, H. Šipová Jungová, G. Emilsson, et al., “Antibody–antigen interaction dynamics revealed by analysis of single-molecule equilibrium fluctuations on individual plasmonic nanoparticle biosensors,” *ACS Nano*, vol. 12, pp. 9958–9965, 2018.
- [5] F. A. A. Nugroho, I. Darmadi, L. Cusinato, et al., “Metal–polymer hybrid nanomaterials for plasmonic ultrafast hydrogen detection,” *Nat. Mater.*, vol. 18, pp. 489–495, 2019.
- [6] A. García-Etxarri, R. Gómez-Medina, L. S. Froufe-Pérez, et al., “Strong magnetic response of submicron silicon particles in the infrared,” *Opt. Express*, vol. 19, pp. 4815–4826, 2011.
- [7] A. I. Kuznetsoc, A. E. Miroshnichenko, Y. Hsing Fu, J. Zhang, and B. Lukyanchuk, “Magnetic light,” *Sci. Rep.*, vol. 2, p. 492, 2012.
- [8] A. B. Evlyukhin, S. M. Novikov, U. Zywietz, et al., “Demonstration of magnetic dipole resonances of dielectric nanospheres in the visible region,” *Nano Lett.*, vol. 12, pp. 3749–3755, 2012.
- [9] A. B. Evlyukhin, C. Reinhardt, and B. N. Chichkov, “Multipole light scattering by nonspherical nanoparticles in the discrete dipole approximation,” *Phys. Rev. B*, vol. 84, p. 235429, 2011.
- [10] I. Staude, A. E. Miroshnichenko, M. Decker, et al., “Tailoring directional scattering through magnetic and electric resonances in subwavelength silicon nanodisks,” *ACS Nano*, vol. 7, pp. 7824–7832, 2013.
- [11] W. Liu and Y. S. Kivshar, “Generalized Kerker effects in nanophotonics and meta-optics [Invited],” *Opt. Express*, vol. 26, pp. 13085–13105, 2018.
- [12] M. Decker, I. Staude, M. Falkner, et al., “High-efficiency dielectric Huygens’ surfaces,” *Adv. Opt. Mater.*, vol. 3, pp. 813–820, 2015.
- [13] P. Moitra, B. A. Slovick, Z. Gang Yu, S. Krishnamurthy, and J. Valentine, “Experimental demonstration of a broadband all-dielectric metamaterial perfect reflector,” *Appl. Phys. Lett.*, vol. 104, p. 171102, 2014.
- [14] V. Giannini, A. I. Fernández-Domínguez, S. C. Heck, and S. A. Maier, “Plasmonic nanoantennas: fundamentals and their use in controlling the radiative properties of nanoemitters,” *Chem. Rev.*, vol. 111, pp. 3888–3912, 2011.
- [15] A. B. Evlyukhin, V. R. Tuz, V. S. Volkov, and B. N. Chichkov, “Bianisotropy for light trapping in all-dielectric metasurfaces,” *Phys. Rev. B*, vol. 101, p. 205415, 2020.
- [16] M. A. Kats, P. Genevet, G. Aoust, et al., “Giant birefringence in optical antenna arrays with widely tailorable optical anisotropy,” *Proc. Natl. Acad. Sci. Unit. States Am.*, vol. 109, pp. 12364–12368, 2012.
- [17] M. B. Ross, M. G. Blaber, and G. C. Schatz, “Using nanoscale and mesoscale anisotropy to engineer the optical response of three-dimensional plasmonic metamaterials,” *Nat. Commun.*, vol. 5, p. 1135, 2014.
- [18] L. Persechini, R. Verre, N. McAlinden, et al., “An analytic approach to modeling the optical response of anisotropic nanoparticle arrays at surfaces and interfaces,” *J. Phys. Condens. Matter*, vol. 26, p. 145302, 2014.
- [19] W. Liu, B. Lei, and A. E. Miroshnichenko, “Q-factor and absorption enhancement for plasmonic anisotropic nanoparticles,” *Opt. Lett.*, vol. 41, pp. 3563–3566, 2016.
- [20] G. A. Ermolaev, D. V. Grudinin, Y. V. Stebunov, et al., “Giant optical anisotropy in transition metal dichalcogenides for next-generation photonics,” *Nat. Commun.*, vol. 12, p. 5789, 2021.
- [21] A. Poddubny, I. Iorsh, P. Belov, and Y. Kivshar, “Hyperbolic metamaterials,” *Nat. Photonics*, vol. 7, pp. 948–957, 2013.
- [22] G. I. López-Morales, M. Li, R. K. Yadav, et al., “Spontaneous emission dynamics of eu3+ ions coupled to hyperbolic metamaterials,” *Appl. Phys. Lett.*, vol. 118, p. 011106, 2021.
- [23] D. J. Roth, A. V. Krasavin, A. Wade, et al., “Spontaneous emission inside a hyperbolic metamaterial waveguide,” *ACS Photonics*, vol. 4, pp. 2513–2521, 2017.
- [24] S. Dai, Q. Ma, T. Andersen, et al., “Subdiffractional focusing and guiding of polaritonic rays in a natural hyperbolic material,” *Nat. Commun.*, vol. 6, p. 2860, 2015.
- [25] G. Hu, J. Shen, C.-W. Qiu, A. Alú, and S. Dai, “Phonon polaritons and hyperbolic response in van der Waals materials,” *Adv. Opt. Mater.*, vol. 8, p. 1901393, 2020.
- [26] J. D. Caldwell, A. V. Kretinin, Y. Chen, et al., “Sub-diffractional volume-confined polaritons in the natural hyperbolic material hexagonal boron nitride,” *Nat. Commun.*, vol. 5, p. 5521, 2014.
- [27] C. Wang, S. Huang, Q. Xing, et al., “Van der Waals thin films of WTe₂ for natural hyperbolic plasmonic surfaces,” *Nat. Commun.*, vol. 11, p. 4592, 2020.
- [28] M.-I. Choe, K.-H. Kim, and J.-H. Wi, “The two-dimensional electrides XONa (X=Mg, Ca) as novel natural hyperbolic materials,” *ChemPhysChem*, vol. 22, pp. 92–98, 2021.
- [29] Z. Liu, H. Lee, Y. Xiong, C. Sun, and X. Zhang, “Far-field optical hyperlens magnifying sub-diffraction-limited objects,” *Science*, vol. 315, pp. 1686, 2007.
- [30] S. S. Kruk, Z. Jing Wong, E. Pshenay-Severin, et al., “Magnetic hyperbolic optical metamaterials,” *Nat. Commun.*, vol. 7, p. 11329, 2016.

- [31] E.D. Palik, Eds. *Handbook of Optical Constants of Solids*, vol. 1, New York, Academic Press, 1985.
- [32] X. Yang, J. Yao, J. Rho, X. Yin, and X. Zhang, “Experimental realization of three-dimensional indefinite cavities at the nanoscale with anomalous scaling laws,” *Nat. Photonics*, vol. 6, pp. 450–454, 2012.
- [33] A. J. Sternbach, S. H. Chae, S. Latini, et al., “Programmable hyperbolic polaritons in van der waals semiconductors,” *Science*, vol. 371, pp. 617–620, 2021.
- [34] S. Wang, X. Cui, C. Jian, et al., “Stacking-engineered heterostructures in transition metal dichalcogenides,” *Adv. Mater.*, vol. 33, p. 2005735, 2021.
- [35] N. Maccaferri, Y. Zhao, T. Isoniemi, et al., “Hyperbolic meta-antennas enable full control of scattering and absorption of light,” *Nano Lett.*, vol. 19, pp. 1851–1859, 2019 [pMID: 30776244].
- [36] K. Wang, P. Filloux, N. Paraire, P. Roca i Cabarrocas, and P. Bulkin, “Two-dimensional photonic crystals by focused-ion-beam etching of multilayer membranes,” *J. Vac. Sci. Technol. B*, vol. 21, pp. 966–969, 2003.
- [37] S. R. K. Chaitanya Indukuri, C. Frydendahl, J. Bar-David, N. Mazurski, and U. Levy, “WS₂ monolayers coupled to hyperbolic metamaterial nanoantennas: broad implications for light–matter-interaction applications,” *ACS Appl. Nano Mater.*, vol. 3, pp. 10226–10233, 2020.
- [38] N. Maccaferri, A. Zilli, T. Isoniemi, et al., “Enhanced nonlinear emission from single multilayered metal–dielectric nanocavities resonating in the near-infrared,” *ACS Photonics*, vol. 8, pp. 512–520, 2021.
- [39] S. Höfer, H. Mutschke, and T. G. Mayerhöfer, “The effect of anisotropy on cross section spectra of uniaxial spherical particles small compared to the wavelength,” *J. Quant. Spectrosc. Radiat. Transf.*, vol. 246, p. 106909, 2020.
- [40] N. Kossowski, P. Y. Chen, Q. J. Wang, P. Genevet, and Y. Sivan, “Scattering by lossy anisotropic scatterers: a modal approach,” *J. Appl. Phys.*, vol. 129, p. 113104, 2021.
- [41] T. Isoniemi, Q. M. Ramasse, N. Maccaferri, G. Strangi, and F. D. Angelis, “Electron energy loss spectroscopy of bright and dark modes in hyperbolic metamaterial nanostructures,” *Adv. Opt. Mater.*, vol. 8, p. 2000277, 2020.
- [42] C. J. Zapata-Rodríguez, J. J. Miret, S. Vuković, and M. R. Belić, “Engineered surface waves in hyperbolic metamaterials,” *Opt. Express*, vol. 21, p. 19113, 2013.
- [43] S. Laref, J. Cao, A. Asaduzzaman, et al., “Size-dependent permittivity and intrinsic optical anisotropy of nanometric gold thin films: a density functional theory study,” *Opt. Express*, vol. 21, pp. 11827–11838, 2013.
- [44] T. Stefaniuk, P. Wróbel, P. Trautman, and T. Szoplik, “Ultrasoft metal nanolayers for plasmonic applications: surface roughness and specific resistivity,” *Appl. Opt.*, vol. 53, pp. B237–B241, 2014.
- [45] A. A. Orlov, P. M. Voroshilov, P. A. Belov, and Y. S. Kivshar, “Engineered optical nonlocality in nanostructured metamaterials,” *Phys. Rev. B*, vol. 84, p. 045424, 2011.
- [46] L. Sun, Z. Li, S. TingLuk, X. Yang, and J. Gao, “Nonlocal effective medium analysis in symmetric metal-dielectric multilayer metamaterials,” *Phys. Rev. B*, vol. 91, p. 195147, 2015.
- [47] S. G. Castillo-López, A. A. Krokhin, N. M. Makarov, and F. Pérez-Rodríguez, “Electrodynamics of superlattices with ultra-thin metal layers: quantum Landau damping and band gaps with nonzero density of states,” *Opt. Mater. Express*, vol. 9, pp. 673–686, 2019.
- [48] A. Andryeuskii, S. V. Zhukovsky, and A. V. Lavrinenko, “Rough metal and dielectric layers make an even better hyperbolic metamaterial absorber,” *Opt. Express*, vol. 22, pp. 14975–14980, 2014.
- [49] S. Kozik, M. A. Binhussain, A. Smirnov, N. Khilo, and V. Agabekov, “Investigation of surface roughness influence on hyperbolic metamaterial performance,” *Adv. Electromagn.*, vol. 3, pp. 6–9, 2014.
- [50] A. Egel and U. Lemmer, “Dipole emission in stratified media with multiple spherical scatterers: enhanced outcoupling from OLEDs,” *J. Quant. Spectrosc. Radiat. Transf.*, vol. 148, pp. 165–176, 2014.
- [51] A. Egel, S. W. Kettlitz, and U. Lemmer, “Efficient evaluation of Sommerfeld integrals for the optical simulation of many scattering particles in planarly layered media,” *J. Opt. Soc. Am. A*, vol. 33, p. 698, 2016.
- [52] A. Egel, K. M. Czajkowski, D. Theobald, K. Ladutenko, A. S. Kuznetsov, and L. Pattelli, “Smuthi: a python package for the simulation of light scattering by multiple particles near or between planar interfaces,” 2021. arXiv:2105.04259 [physics.optics].
- [53] A. D. Kiselev, V. Yu Reshetnyak, and T. J. Sluckin, “Light scattering by optically anisotropic scatterers: T-matrix theory for radial and uniform anisotropies,” *Phys. Rev. E*, vol. 65, p. 056609, 2002.
- [54] A. Doicu, T. Wriedt, and Y. A. Eremin, *Light Scattering by Systems of Particles Null-Field Method with Discrete Sources: Theory and Programs*, Berlin, Springer, 2014.
- [55] C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles*, Weinheim, Wiley, 1998.
- [56] $\Psi_1(\epsilon_\perp, \epsilon_\parallel) = (-1.5 + 0.25\epsilon_\perp)\epsilon_\perp^3 + \epsilon_\perp^2(-4.95 + \epsilon_\perp(1.08 + \epsilon_\perp))\epsilon_\parallel + \epsilon_\perp(-4.1 + \epsilon_\perp(1.5 + 3.3\epsilon_\perp))\epsilon_\parallel^2 + \epsilon_\perp(0.68 + 2.73\epsilon_\perp)\epsilon_\parallel^3$.
- [57] $\Psi_3(\epsilon_\perp, \epsilon_\parallel) = -16.3\epsilon_\perp^3 + \epsilon_\perp^2(-49 - 43.27\epsilon_\parallel) + \epsilon_\perp(-81 - 27\epsilon_\parallel)\epsilon_\parallel$.
- [58] A. Moroz, “Depolarization field of spheroidal particles,” *J. Opt. Soc. Am. B*, vol. 26, pp. 517–527, 2009.
- [59] F. M. Schulz, K. Stamnes, and J. J. Stamnes, “Point-group symmetries in electromagnetic scattering,” *J. Opt. Soc. Am. A*, vol. 16, p. 853, 1999.
- [60] S. Gladyshev, K. Frizyuk, and A. Bogdanov, “Symmetry analysis and multipole classification of eigenmodes in electromagnetic resonators for engineering their optical properties,” *Phys. Rev. B*, vol. 102, p. 075103, 2020.
- [61] Z. Xiong, Q. Yang, W. Chen, Z. Wang, J. X. W. Liu, and Y. Chen, “On the constraints of electromagnetic multipoles for symmetric scatterers: eigenmode analysis,” *Opt. Express*, vol. 28, pp. 3073–3085, 2020.
- [62] K. M. Czajkowski, M. Bancerek, and T. J. Antosiewicz, “Multipole analysis of substrate-supported dielectric nanoresonator metasurfaces via the t-matrix method,” *Phys. Rev. B*, vol. 102, p. 085431, 2020.
- [63] T. J. Antosiewicz, S. P. Apell, C. Wadell, and C. Langhammer, “Absorption enhancement in lossy transition metal elements of plasmonic nanosandwiches,” *J. Phys. Chem. C*, vol. 116, pp. 20522–20529, 2012.

- [64] A. R. Beal, H. P. Hyghes, and W. Y. Liang, “The reflectivity spectra of some group VA transition metal dichalcogenides,” *J. Phys. C*, vol. 8, pp. 4236–4248, 1975.
- [65] Q. Yan-Bin, Li. Yan-Ling, Z. Guo-Hua, Z. Zhi, and Q. Xiao-Ying, “Anisotropic properties of TaS₂,” *Chin. Phys.*, vol. 16, pp. 3809–3814, 2007.
- [66] T. Stefaniuk, P. Wróbel, E. Górecka, and T. Szoplik, “Optimum deposition conditions of ultrasmooth silver nanolayers,” *Nanoscale Res. Lett.*, vol. 9, p. 153, 2014.
- [67] R. Verre, L. Shao, N. Odebo Länk, et al., “Metasurfaces and colloidal suspensions composed of 3d chiral si nanoresonators,” *Adv. Mater.*, vol. 29, p. 1701352, 2017.
- [68] A. Lehmuskero, M. Kuittinen, and P. Vahimaa, “Refractive index and extinction coefficient dependence of thin al and ir films on deposition technique and thickness,” *Opt Express*, vol. 15, pp. 10744–10752, 2007.
- [69] A. Orlov, I. Iorsh, P. Belov, and Y. Kivshar, “Complex band structure of nanostructured metal-dielectric metamaterials,” *Opt. Express*, vol. 21, pp. 1593–1598, 2013.
- [70] R. C. Monreal, T. J. Antosiewicz, and S. P. Apell, “Competition between surface screening and size quantization for surface plasmons in nanoparticles,” *New J. Phys.*, vol. 15, p. 083044, 2013.
- [71] R. C. Monreal, S. P. Apell, and T. J. Antosiewicz, “Surface scattering contribution to the plasmon width in embedded ag nanospheres,” *Opt Express*, vol. 22, pp. 24994–25004, 2014.
- [72] B. Munkhbat, D. G. Baranov, A. Bisht, et al., “Electrical control of hybrid monolayer tungsten disulfide–plasmonic nanoantenna light–matter states at cryogenic and room temperatures,” *ACS Nano*, vol. 14, pp. 1196–1206, 2020.
- [73] C. Díaz-Aviñó, M. Naserpour, and C. J. Zapata-Rodríguez, “Tunable scattering cancellation of light using anisotropic cylindrical cavities,” *Plasmonics*, vol. 12, pp. 675–683, 2016.
- [74] S. R. K. Chaitanya Indukuri, J. Bar-David, N. Mazurski, and U. Levy, “Ultrasmall mode volume hyperbolic nanocavities for enhanced light–matter interaction at the nanoscale,” *ACS Nano*, vol. 13, pp. 11770–11780, 2019.
- [75] K. V. Sreekanth, Y. Alapan, M. El Kabbash, et al., “Extreme sensitivity biosensing platform based on hyperbolic metamaterials,” *Nat. Mater.*, vol. 15, pp. 621–627, 2016.

Supplementary Material: The online version of this article offers supplementary material (<https://doi.org/10.1515/nanoph-2021-0247>).