Polarization-dependent mode coupling in hyperbolic nanospheres

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Hyperbolic materials offer a 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 of 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 resonance 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 strongly scattering or absorbing modes. These observations assert that hyperbolic nanoantennas offer a promising route towards novel light-matter interaction regimes.

I. 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 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 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 restrahlen 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 ma-
II. 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 Au-SiO$_2$/Al$_2$O$_3$/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 $\epsilon$ by combining a dispersionless dielectric with refractive index $n_d$ and silver [31]. The nanoresonators, here spherical in shape, are schematically represented in Fig. 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 $\epsilon_m$ and $\epsilon_d = n_d^2$.

For a metal fill factor $f_m = 0.5$ the exemplary dispersion is plotted in Fig. 1b, which illustrates the presence of two types of isofrequency surfaces [21] with either two (below $\sim$3.4 eV) or one ($\sim$3.4–3.8 eV) permittivity tensor elements being negative. For a larger $f_m$, $\epsilon$ 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 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 the need in stacking transition metal dichalcogenide (TMD) heterostructures [34] and, 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 [33] or etch nanostructures in prepared multilayers [36]. Indeed, optical type II hyperbolic nanoantennas have been recently shown to exhibit simultaneously radiative and non-radiative 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.
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 a given.

The optical spectra (from FDTD) for three unique plane wave incident/polarization combinations are then plotted in Fig. 2 for a sphere with radius $r = 50 \text{ nm}$, $f_m = 0.5$, and 4 nm layers and compared to T-matrix calculations (open-source code SMUHTH [50–52]) for an effective hyperbolic permittivity. The agreement between the scattering and absorption spectra, Fig. 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 Fig. 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 Fig. 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 Fig. 2a,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 $\sim 2.5 \text{ eV}$, which shows up in scattering and absorption, and a second resonance at $\sim 1.7 \text{ eV}$, which consists of a 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 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 (Fig. 2c,d) or parallel to the anisotropy axis (Fig. 2e,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 Fig. 2a,b. In the latter case in Fig. 2e,f one sees a strong resonance $\sim 3.6 \text{ eV}$ which consists of the ED, MD, and EQ. Additionally, a second absorptive resonance (with practically no scattering) is seen at $\sim 1.7 \text{ eV}$, which is qualitatively similar to the one at the same energy in Fig. 2a,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.

### A. 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 non-radiative 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 [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 $(\tau, m, l)$. Here $\tau$ 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 2x2 blocks given by equation

$$T^{\tau_1,\tau_2}_{m_1,l_1,m_2,l_2} = -Q^{\tau_1,\tau_2}_{m_1,l_1,m_2,l_2} [Q^{\tau_2,\tau_1}_{m_1,l_1,m_2,l_2}]^{-1}.$$

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

$$Q^{3,1,1}_{m_1,l_1,m_2,l_2} = \frac{ix^2}{\pi} \int [m_r (\hat{r} \times X^h_{m_1,l_1}) \cdot M_{m_2,l_2}^3 + (\hat{r} \times X^e_{m_1,l_1}) \cdot N_{m_2,l_2}^3] dS,$$

where $M$ and $N$ are the vector spherical wave functions, $\hat{r}$ is the unit vector normal to the sphere surface, $X^e$ and $X^h$ are internal modes of the anisotropic sphere. Changing the first $Q^3$ matrix index leads to exchange of $M$ for $N$ and vice versa. Changing the second index leads to replacing of $X$ internal modes to $Y$ internal modes as shown in [54]. Expressions for $Q^1$ are similar, but with $(M^3, N^3)$ replaced by $(M^1, N^1)$. The internal modes of the anisotropic sphere are described in terms of plane waves, which are parametrized by angles $(\alpha, \beta)$ 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,

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

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:

\[ T_{1,1,1,1}^{1,1} = -\frac{2}{3}ix^3 \frac{(-1 + \epsilon_{\perp})}{2 + \epsilon_{\perp}} \]  
\[ T_{0,1,0,1}^{1,1} = -\frac{2}{3}ix^3 \frac{(-1 + \epsilon_{\parallel})}{2 + \epsilon_{\parallel}} \]  

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

\[ \epsilon_{\perp} = -2 \quad \text{and} \quad \epsilon_{\parallel} = -2. \]  

Equation (6a) corresponds to the incident field with \( E_{\perp} \) for both \( k_{\parallel} \) and \( k_{\perp} \), cf. Fig. 2a-d and eq. (6b) for \( E_{\parallel} \) and \( k_{\perp} \), cf. Fig. 2e,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 Fig. 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, Fig. 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

\[
T_{1, 1, 1}^{0, 0} = 0.725 i x^3 \frac{\Psi_1(\epsilon_{\|}, \epsilon_\perp)}{(\epsilon_{\perp} + 1.65 \epsilon_{\|}) \Psi_3(\epsilon_{\perp}, \epsilon_{\|})}, \tag{7}
\]

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

The magnetic resonance observed in Fig. 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

\[
\epsilon_\perp + 1.65 \epsilon_{\|} = 0. \tag{8}
\]

This equation, qualitatively corresponding to the Fröhlich 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 \(|T|^2 / \text{Re}(T)|^2\). For small spheres the magnetic moment is proportional to \(x^3\) according to eq. 7, while the electric moment is proportional to \(x^3\) (Eqs. 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 Fig. 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 Fig. 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 Fig. 3b.

Furthermore, depolarization leads to a shift of the resonance energy. For the smallest nanoparticle size \((r = 5 \text{ nm})\) considered in Figs. 3c-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.

### B. 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 non-zero 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 $m$

$$T_{m_{1},l_{1},m_{2}}^{\tau_{1},\tau_{2}} = \delta_{m_{1},m_{2}}T_{m_{1},m_{2}}^{\tau_{1},\tau_{2}}.$$  \hspace{1cm} (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 $-m$

$$T_{m_{1},l_{1},m_{2}}^{\tau_{1},\tau_{2}} = (-1)^{\tau_{2}+\tau_{2}}T_{m_{1},m_{2},l_{1}}^{\tau_{1},\tau_{2}}.$$  \hspace{1cm} (10)

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

$$T_{m_{1},l_{1},m_{2}}^{\tau_{1},\tau_{2}} T_{m_{2},l_{2},m_{1}}^{\tau_{2},\tau_{2}} = T_{m_{1},l_{1},m_{2}}^{\tau_{1},\tau_{2}} T_{m_{2},l_{2},m_{1}}^{\tau_{2},\tau_{2}}.$$  \hspace{1cm} (11)

Finally, the presence of a horizontal plane of reflection imposes

$$T_{m_{1},l_{1},m_{2}}^{\tau_{1},\tau_{2}} = 0,$$  \hspace{1cm} (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 Fig. 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 parallel-parallel, perpendicular-perpendicular and parallel-perpendicular etc.

Simultaneously, the T-matrix has non-zero 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.

### C. 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 $\sigma \propto \text{Re}(a^{\prime}T\mathbf{a})$, where $\mathbf{a}$ is a vector of the initial field coefficients in the $(\tau, m, l)$ basis. To facilitate analysis of the mode structure, it is convenient to recast extinction of a given $(\tau, m, l)$ mode into two parts. The first is connected with pure mode excitation and the second results from coupling between modes with various $\tau$, $m$ and $l$

$$\sigma_{m,l}^{\tau} \propto \text{Re} \left[ |a_{m,l}^{\tau}|^2 \sum_{m',l',\tau'} \frac{T_{m,l,m,l}^{\tau,\tau'} a_{m,l}^{\tau'} + a_{m,l}^{\tau'} * \sum_{m',l',\tau'} T_{m,l,m,l}^{\tau,\tau'} a_{m',l'}^{\tau} \right].$$  \hspace{1cm} (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. $a_{m,l}^{\tau}$ and $a_{m',l'}^{\tau'}$ product.

Now we focus on plane wave illumination with initial field coefficients $a_{P,m,l}^{\tau}$ where we add the index $P$ to denote the wave’s polarization. For normal incidence along the optical axis, $\mathbf{E}_{\parallel}$ with $k_{\parallel}$ (equivalent to $\mathbf{T}_{\parallel}$ 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]

$$a_{\text{TE}_{\parallel},l,0}^{0} = i^{l+1} \sqrt{2l+1},$$  \hspace{1cm} (14)

$$a_{\text{TE}_{\parallel},l,0}^{1} = i^{l} \sqrt{2l+1} \frac{(-1)^{l}}{i^{l}}.$$  \hspace{1cm} (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 $\mathbf{T}_{\perp}$ and $\mathbf{E}_{\perp}$ polarizations. In the first case for TM with $k_{\perp}$ and $E_{\parallel}$ we have

$$a_{\text{TM}_{\perp},l,m}^{0} = \begin{cases} \alpha_{m,l}^{0} & l + |m| \text{ even} \\ \frac{(l+|m|)!(l-|m|+1)!}{(l+1-|m|)!} \alpha_{m,l}^{1} & l + |m| \text{ odd} \end{cases}$$  \hspace{1cm} (16)

$$a_{\text{TM}_{\perp},l,m}^{1} = \begin{cases} 0 & l + |m| \text{ even} \\ \frac{(l+|m|)!(l-|m|+1)!}{(l+1-|m|)!} \alpha_{m,l}^{0} & l + |m| \text{ odd} \end{cases}$$  \hspace{1cm} (17)

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

$$a_{\text{TE}_{\perp},l,m}^{0} = \begin{cases} -im \frac{(l+|m|)!}{(l-|m|)!} \alpha_{m,l}^{1} & l + |m| \text{ even} \\ 0 & l + |m| \text{ odd} \end{cases}$$  \hspace{1cm} (18)

$$a_{\text{TE}_{\perp},l,m}^{1} = \begin{cases} 0 & l + |m| \text{ even} \\ \frac{(l+|m|)!}{(l+1-|m|)!} \alpha_{m,l}^{0} & l + |m| \text{ odd} \end{cases}$$  \hspace{1cm} (19)

The above used $\alpha_{m,l}$ 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
For the three unique polarizations is shown in Fig. 4. Moreover, only an incident field with $k_\parallel$ (TE$_\parallel$/TM$_\parallel$) with an azimuthal mode $m = \pm 1$ can be expressed in relation to normal incidence ($k_\parallel$) as

$$a^r_{P_{\parallel,\pm 1, t}} = (-1)^{l-1} a^r_{P_{\parallel,\pm 1, t}}. \quad (21)$$

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 Fig. 5 and in the SI for the three unique incidence/polarization cases.

For polarization $E_\perp$ and irrespective of $k$ direction, a
dominant ED is always present and of the same amplitude at a given $r$, cf. Fig. 5ab, 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 [eq. (15), eq. (19)] and ED is negligibly coupled to other resonances. However, the higher electric multipoles are only visible in the plots in Fig. 5b with $k_\parallel$, 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 Fig. 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 Fig. 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 $k_\parallel$ (Fig. 5a), which is weakly radiative for small $r$ as discussed above. Neither the MD nor other magnetic modes are present in Fig. 5b (TE$_\perp$), when the spectra of a HNS are reminiscent of an isotropic plasmonic nanosphere for all radii (and \(\lesssim 3.5\) eV). Magnetic modes are, however, present in Fig. 5c (TM$_\perp$), specifically the magnetic one is found at the same spectral location as in Fig. 5a. However, its amplitude is much weaker due to destructive interference from the coupled EQ. Conversely, in Fig. 5a the MD and EQ interfere constructively (cf. Fig. 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 Fig. 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 Fig. 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 Figs. 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, this particles’ extinction may turn negative, implying that it effectively returns more energy to the electromagnetic field than it receives 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 a HNS (cf. Fig. 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 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.

D. 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 Fig. 1cd, varying $f_m$ allows for significant tunability from a uniaxial dielectric through a type I or II hyperbolic material to a uniaxial metal. In Fig. 6 we present how the extinction spectrum and its multipole decomposition evolve with $f_m$ for a 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 Fig. 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 and EO resonances appear for TEM and TM$_\perp$ polarizations when the material becomes a uniaxial metal.

The MD response is, however, quite strong and clearly
seen in for Fig. 6ac. 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 Fig. 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 \( \epsilon_m(\omega) = \epsilon_\infty - \omega_p^2/(\omega^2 + i\gamma\omega) \) 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

\[
\omega_{res}^{MD} \approx \frac{\omega_p \sqrt{F_m}}{\sqrt{\epsilon_\infty F_m - \epsilon_d(1.3 + F_m + 1.6\sqrt{0.7 + F_m})}},
\]

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 symmetric, crescent-shaped of the MD resonance confirms 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 polarizations. Both approach the well-established result for a quasi-static sphere \( \omega_{res} = \omega_p \sqrt{f_m} \) in the limit of purely metallic particles and confirm the earlier observation on the quasi-linear 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 \( \omega_p \) 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.

### E. 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 hyperbolic material were not common. However, with the rise of extensive research on vdW materials hyperbolic materials can easily be found in the literature. Examples include hBN with two restrahlen bands [26] or materials with Drude-like dispersion for the in-plane components, such as considered here, TaS\(_2\) or TaSe\(_2\) [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 multi-layers 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 \( \sim 1 \) eV. As we show in Fig. 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 (Fig. 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 (Fig. 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 (Fig. 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 are not expected to play a role in our case. 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 modelled 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].

III. 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_{⊥} 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 multipole 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_{⊥} 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.

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