Dependence of optical modes in hyperbolic metamaterials on surrounding medium

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***Abstract*—The optical response of an hyperbolic nanostructures is governed by the interplay of two resonances: an electric dipole (ED) and a magnetic dipole coupled with an electric quadrupole (MD-EQ). Their spectral positions and amplitudes are influenced by the medium surrounding the nanoresonator. Here, I present a derivation of resonance conditions dependent on dielectric permittivity of external medium for ED and MD-EQ modes in a quasistatic hyperbolic nanosphere (HNS).**

***Index Terms*—Hyperbolic metamaterials, optical sensors, refractometric sensitivity, resonance conditions**

I. Introduction

Hyperbolic metamaterials (HMMs) significantly broaden the scope of material engineering, opening up new possibilities for controlling the propagation of light [1]. Their unusual optical properties stem from a specific type of anisotropy – in the permittivity tensor of uniaxial HMMs two diagonal elements are of opposite sign to the other one. This anisotropy results in a hyperbolic dispersion relation, which in turn is responsible for the negative refraction of light [2]. HMMs have also been implemented in super-resolution imaging [3], enhanced spontaneous emission [4], or optical biosensors [5].

Arranging subwavelength elements of metal and dielectric materials in artificial HMMs combines the properties of both constituent materials within a single structure. HMMs support both electric and magnetic resonances, resulting in a rich modal optical response. Here, I consider a hyperbolic nanosphere (HNS) made of an Ag:SiO_2 multilayer (see Fig. 1a). In my calculations the dielectric permittivity of silver is described by the Drude model ($\epsilon_m = \epsilon_\infty - \omega_p^2/(\omega^2 + i\gamma\omega)$, with ϵ_∞ = 3.37, ω_p = 9.83 eV, γ = 0.23 eV) and the dielectric is non-dispersive ($\epsilon_d = 2.25$) (see Fig. 1b for the components of the effective medium permittivity tensor). Its extinction spectrum is defined by a strongly scattering electric dipole (ED) resonance and an absorptive magnetic dipole (MD) coupled with an electric quadrupole (EQ) mode (see Fig. 1 c–e).

The excitation conditions of these resonances are influenced by numerous factors, such as the size, shape, and fill factor of the metal [6]. Both electric and magnetic resonances in HMMs are affected also by the external medium with its dielectric permittivity ϵ_s. In general, relating the inherent properties of the nanoparticle and the permittivity of the surrounding medium in a simple resonance condition would be beneficial for a better understanding of excited resonances and the engineering of optical materials.

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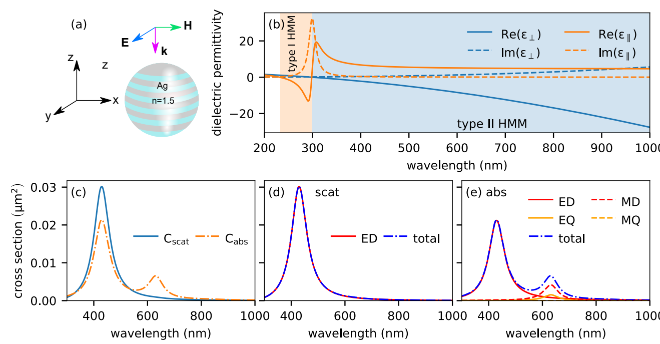

Fig. 1. (a) Hyperbolic nanosphere. (b) Dispersion relation graph of an effective medium Ag:SiO_2 multilayer (with metal fill factor $ff_m = 0.5$). (c) Scattering and absorption cross sections of an Ag:SiO_2 nanosphere with 40 nm radius. (d) Scattering and (e) absorption multipole decomposition.

Caldwell et al. [7] directly tied the resonance energy to the material and geometry of hyperbolic nanocones. They derived resonance condition based on the quantization rule of the geometrical aspect ratio in disk-like particles. Additionally, the resonance condition derived in [8] states that the MD resonance occurs in HMMs when $\epsilon_\perp = -1.65\epsilon_\parallel$. Here, $\epsilon_\perp(\epsilon_\parallel)$ is the effective dielectric permittivity in the direction perpendicular (parallel) to the anisotropy axis. However, both resonance conditions are limited by (i) the subwavelength size of the resonators and (ii) the omission of the effect of the surrounding external medium. Although the plasmonic ED resonance is determined by the Fröhlich condition, $\epsilon_m = -2\epsilon_s$, an analogous resonance condition for MD has been lacking.

II. Results and Discussion

In our work we have derived resonance conditions dependent on the dielectric permittivity of the surrounding medium for a quasistatic hyperbolic nanosphere (HNS). We applied the Null-Field Method algorithm [9] in the quasistatic approximation ($x \equiv kr \ll 1$, where r is the sphere's radius and k is the wavenumber in the external medium). By expanding the internal fields into plane waves and solving the boundary problem, we calculated the transition matrix (T-matrix) of a quasistatic HNS. The T-matrix explicitly describes the multipolar optical response of a single scatterer. Therefore, by expanding the T-matrix components corresponding to the relevant multipoles into a Taylor series, we obtained the T-matrix in simple polynomial form for the ED [10]:

$$T^{ED} = -\frac{2}{3} i x^3 \frac{\epsilon_\perp - \epsilon_s}{\epsilon_\perp + 2\epsilon_s} \quad . \tag{1}$$

and the MD-EQ mode (for simplicity called MD):

$$T^{MD} = i x^5 \frac{\epsilon_s^{3/2}(6\epsilon_s^2 - 4\epsilon_\perp \epsilon_\parallel - \epsilon_s(\epsilon_\perp + \epsilon_\parallel))}{90(\epsilon_\perp + \epsilon_\parallel + 3\epsilon_s)}. \tag{2}$$

In case of the ED, the result is consistent with Mie scattering theory and leads to the Fröhlich condition for plasmonic resonances:

$$\epsilon_\perp = -2\epsilon_s \tag{3}$$

The ED is a purely plasmonic resonance that is not coupled to any other mode. Contrarily, the MD resonance only occurs in a HMM when coupled to the EQ. Thus, an EQ must be accounted for in the T-matrix calculations. From the pole of the denominator in (2), we derived a material-dependent resonance condition for the MD:

$$\epsilon_\perp = -\epsilon_\parallel - 3\epsilon_s. \tag{4}$$

The above condition explicitly links the dielectric permittivity of a HNS at resonance with the permittivity of the surrounding medium. Moreover, it is satisfied only by a HMM, in which $\epsilon_\perp$ and $\epsilon_\parallel$ have opposite signs. However, we did not assume any particular type of anisotropic dispersion in the T-matrix calculations. Therefore, (3) and (4) are applicable to both type I and type II hyperbolic dispersion.

We then verify the accuracy of conditions (3) and (4) by comparing their predictions with full T-matrix calculations for an Ag:SiO_2 hyperbolic nanosphere (see Fig. 1a) with an increasing radius from 5 to 50 nm in 5 nm increments. Numerical simulations were performed using an open-access software called SMUTHI [11]. The nanosphere is surrounded by a medium with dielectric permittivity ϵ_s increasing from 1 to 2.25 (see Fig. 2). The resonant wavelengths of the ED in a quasistatic HNS (5 nm in radius) are in good agreement with the quasistatic predictions from (3). Contrary to the ED, the spectral positions of the MD resonance predicted by (4) follow simulations' results for a 15 nm nanosphere. The discrepancy between the resonance predictions and the full T-matrix calculations stems from the neglect of losses in the derived resonance conditions. Equations (3) and (4) only account for the real part of permittivity. The MD is a predominantly absorptive resonance; therefore, neglecting the material losses causes the predicted resonance wavelength to redshift. Naturally, since the resonance conditions were derived under the quasistatic approximation, the calculated resonance positions of both the ED and the MD further deviate from the quasistatic predictions as the particle radius increases. Overall, however, the simulation results are in good agreement with the resonance predictions for a quasistatic HNS.

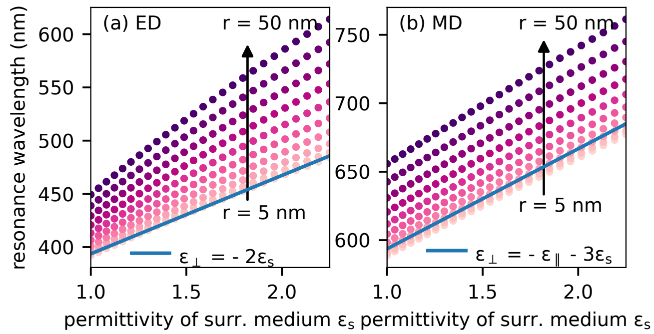

Fig. 2. (a) ED and (b) MD resonance wavelengths vs. permittivity of surrounding medium for hyperbolic nanospheres with radius ranging from 5 nm to 50 nm every 5 nm. Comparison of full T-matrix calculations (circles) with quasistatic predictions (lines).

III. Conclusion

In conclusion, the optical response of hyperbolic nanoparticles is determined by ED and MD-EQ resonances. The excitation conditions of these modes are related to the permittivity of the surrounding medium, as we demonstrated in our derived resonance conditions. The predictions of these resonance conditions are in good agreement with the simulation results for a quasistatic HNS.

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