Optical magnetic response without metamaterials

Cite as: APL Photon. 6, 071303 (2021); doi: 10.1063/5.0054752
Submitted: 21 April 2021 • Accepted: 5 July 2021 •
Published Online: 15 July 2021

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ABSTRACT

One of the main achievements of metamaterials research has been the development of structured matter exhibiting optical magnetism: first, in an array of microwave split-ring resonators and, soon after, in plasmonic and dielectric metamaterials at THz to visible frequencies. We show here that metamaterial structuring is not necessary to achieve optical magnetic response. Indeed, such a response is an essential characteristic of homogeneous dielectric thin films—Fabry–Pérot resonances, for example, depend on interference among electromagnetic multipoles including the magnetic dipole.

The observation of a magnetic response at high (optical) frequencies was a fundamental step in electromagnetism, enabled by metamaterials—artificial media periodically structured on the subwavelength scale.[1,2] This appeared to contradict the long-held idea that magnetization and permeability lose their meaning at high frequencies and that "optical magnetism" did not exist.[3] However, this (Landau–Lifshitz) argument does not hold in structured plasmonic metals and high refractive index dielectrics.[4] Historically, the rapid rise of the metamaterials research field was driven by opportunities arising through the realization of materials manifesting optical magnetism, which does not exist in nature. It is necessary for negative refraction and in turn for "superlenses,"[5] certain forms of "cloaking,"[6] and various beam-steering/wave-guiding applications. It is also of fundamental importance and increasing interest in the context of electromagnetic nonreciprocity and opto-magnetic devices.[7–9]

In metamaterials comprising metallic split-ring metamolecules, the optical magnetic response is typically underpinned by oscillating conduction current loops induced by the electric component of the incident light field.[1,2,13] In dielectric metamaterials or individual nanoparticles, it is associated with loops of displacement current induced in the nanostructured dielectric, again by the incident electric field.[14–16] We show here that an optical magnetic response is in fact a characteristic feature of homogeneous, unstructured dielectric layers of sub-wavelength thickness. Well-known thin film properties, such as Fabry–Pérot (FP) interference resonances, cannot be explained without the magnetic contribution. The vanishing of reflectivity at the fundamental FP resonance is a consequence of destructive interference among electric dipole (ED), magnetic dipole, and electric quadrupole (EQ) contributions. The second-order FP resonance is associated with higher order multipoles: the electric octupole (EO), magnetic quadrupole (MQ), and toroidal dipole (TD) exhibit resonances at this point, while the electric dipole vanishes.

The multipolar nature of the optical response of a dielectric layer is a universal feature of thin films, and it is most pronounced in high refractive index media. Here, we numerically solve Maxwell's equations for light propagation through the layer (using the finite element method in COMSOL MultiPhysics) and evaluate the displacement currents induced by the incident wave [Figs. 1(a)–1(d)]. (The problem can also be solved analytically—see the supplementary material.) From there, we compute the different multipole contributions to light scattering [Fig. 2(b)] using the standard methodology employed in metamaterials research.[17] The

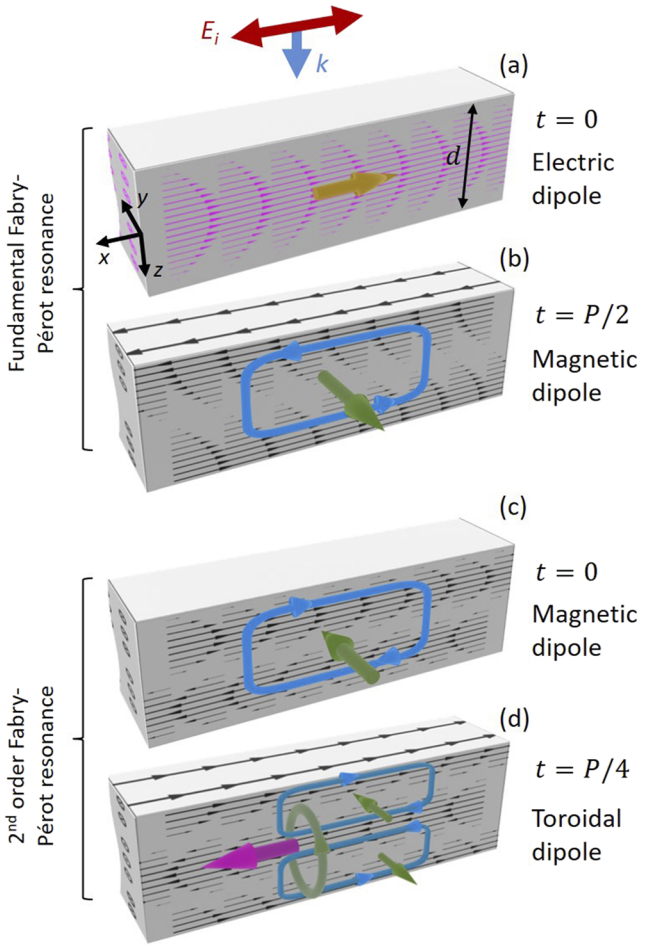

FIG. 1. Electromagnetic multipoles induced in a thin dielectric film by a normally incident plane wave at the [(a) and (b)] fundamental and [(c) and (d)] second-order Fabry–Pérot resonance wavelengths at moments of time t separated by a half-period ($P/2$) for the fundamental and a quarter-period ($P/4$) for the second-order resonance. Magenta arrows in (a) relate to the electric displacement field amplitude and direction; black arrows in (b)–(d) to the displacement current. Overlaid 3D arrows schematically show the significant multipoles in each case: electric, magnetic, and toroidal dipoles in gold, green, and purple; displacement and poloidal current loops in blue and green, respectively.

accuracy of the multipole decomposition is verified by comparing transmission and reflection spectra obtained via numerical Maxwell solving with spectra calculated as the sum of contributions from the multipoles. Here, inclusion of multipole terms up to the magnetic octupole[18] is sufficient to describe the macroscopic optical properties of the film with accuracy better than 1% [Fig. 2(a)]. Our analysis shows that the optical properties of a subwavelength thickness dielectric layer are not solely of electric dipolar nature but formed by a combination of multipole excitations [Fig. 2(b)]. Indeed, under certain resonant conditions, the electric dipolar response vanishes. The magnetic dipolar contribution is prominent at all wavelengths (resonant or otherwise), and its omission leads to significant errors (exceeding 50%) in the reconstruction of

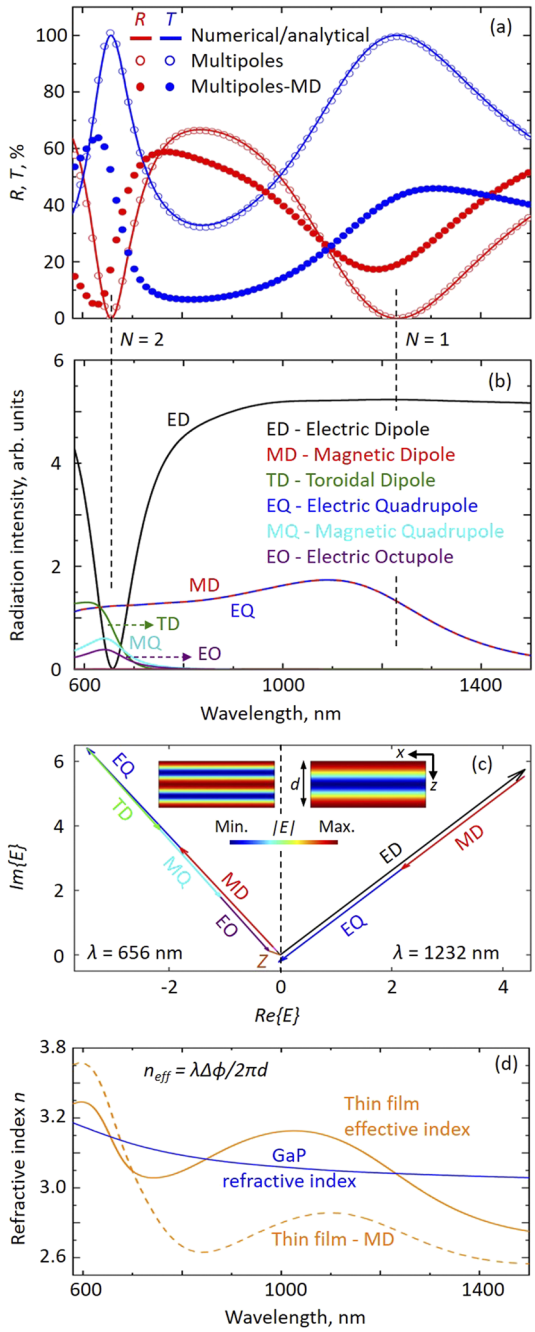

FIG. 2. (a) Spectral dependence of transmission (T, blue) and reflection (R, red) of a 200 nm GaP film in vacuum at normal incidence. Solid lines are identically obtained by numerically solving Maxwell's equations or analytically. Hollow circles are derived from multipole scattering reconstructions. Solid circles are the same but excluding the magnetic dipole contribution. (b) Spectral dependence of light intensity radiated in the reflection direction by the six leading multipoles. (c) Vector plot of complex electric field multipole components at the 656 and 1232 nm FP resonant wavelengths. The insets show corresponding cross-sectional electric field distributions. (d) Dispersion of the GaP film's effective index ($\Delta\varphi$ = phase delay in transmission) and GaP's bulk refractive index. The dashed line shows the film's effective index without accounting for the magnetic dipole contribution.

reflection and transmission spectra of the layer, as illustrated in Fig. 2(a).

FP resonances are observed when the optical thickness of a layer is equal to an integer number N of standing wave periods: $N\lambda/2 = n(\lambda)d$, where d is the physical thickness of the layer and $n(\lambda)$ is its wavelength-dependent refractive index. Here, we consider a 200 nm thick GaP layer that presents FP resonances in the visible to near-IR spectral range, at 1232 nm (N = 1) and 656 nm (N = 2), where transmissivity approaches unity and reflectivity vanishes [Fig. 2(a)].

At the fundamental (N = 1) resonance, the response is dominated by electric dipole (ED) scattering, with magnetic dipole (MD) and electric quadrupole (EQ) contributions [Fig. 2(b)]. The induced ED arises from strong in-phase displacement fields $\vec{D} = \varepsilon\vec{E}$, where ε is the dielectric permittivity and $\vec{E}$ is the electric field inside the dielectric. $\vec{D}$ and $\vec{E}$ are unidirectionally oriented across the film thickness [magenta arrows in Fig. 1(a)]. The induced MD emerges from opposingly directed displacement currents $\vec{J_D} = i\omega\varepsilon_0(\varepsilon - 1)\vec{E}$ [black arrows in Fig. 1(b)] at the top/bottom of the layer, forming a magnetic moment current loop in the xz plane. The ED oscillates with a half period (P/2) phase difference from the MD/EQ, whereby their radiated fields interfere destructively. This is further illustrated in Fig. 2(c), which presents the real and imaginary parts of the multipolar fields scattered (reflected) in the backward ($+z$) direction. The MD and EQ contributions are each equal to around half that of the ED but oscillate in antiphase with the latter.

At the second (N = 2) FP resonance, ED emission vanishes but, alongside the MD and EQ, we observe contributions from higher multipoles including the electric octupole (EO), magnetic quadrupole (MQ), and toroidal dipole (TD) [Fig. 2(b)]. The induced MD again emerges from opposing displacement currents [black arrows in Fig. 1(c)] at the top/bottom of the layer, forming a magnetic moment current loop in the xz plane. The TD response is derived from two counter-rotating displacement current loops [Fig. 1(d)] that give rise to a poloidal current loop in the yz plane. Fields radiated by the TD exhibit a quarter cycle (P/4) phase difference with respect to the TD moment and are thus in antiphase with the MD. The MD and EQ emission components are again (as at N = 1) in-phase and of comparable amplitude, now being the two largest components. Reflectance is canceled by their destructive interference with fields radiated by the EO, MQ, and TD, as illustrated in Fig. 2(c) (here, the vector Z represents the collective contribution of still higher order terms).

Figure 2(d) shows the dispersion of the GaP film's effective refractive index, defined as $n_{eff} = \lambda\Delta\varphi/2\pi d$, where $\Delta\varphi$ is the phase delay in transmission. By virtue of interference between the electric and magnetic responses, this can be higher or lower than the material's volume refractive index. Here again, as for the reflectivity and transmission spectra in Fig. 2(a), the essential role of the magnetic component of optical response is clearly illustrated by the fact that omission of the MD from the multipolar reconstruction leads to significant errors in the value and dispersion of the thin film effective index. The effective enhancement/suppression of the index through multipolar interference in the present case resembles the mechanism whereby a negative refractive index is achieved in metamaterials via the simultaneous presence of (engineered) electric and magnetic responses. The regime of suppressed reflectivity may also be compared with that of (i) optical anapole metamaterials,[19,20] in which it derives from interference between electric and toroidal dipoles, and (ii) Huygens metasurfaces, wherein it arises through interference of electric and magnetic responses.[21,22] In thin unstructured dielectric layers, reflectivity suppression is achieved without lateral (metamaterial) structuring, while, nonetheless, being related to multipolar interference. We also note that the magnetic and generally multipolar nature of a dielectric film's response has much in common with the enhanced magnetic response observed in thin ($<\lambda$) films in a standing wave.[23]

In conclusion, we would emphatically agree with Monticone and Alù[24] that "artificial optical magnetism ... represents a quintessential example of how new fundamental material properties, previously thought to be strictly unavailable, can truly be realized by the engineered arrangement of elements at the nanoscale," and have shown here that it can be achieved by simple confinement of a material in a subwavelength layer. The complex structure of multipole fields in a thin layer of dielectric can be exploited for coupling of electromagnetic radiation to matter through the excitation of high-order multipolar atomic transitions in constituent atoms,[10] and for the exploitation of magnetic dipole (MD) transitions at optical frequencies, for instance, in rare earths as laser gain media[11] and quantum qubit applications.[12]

See the supplementary material for the details of numerical and analytical calculation methods.

This work was supported by the Engineering and Physical Sciences Research Council, UK (Grant No. EP/M009122/1 and EP/T02643X/1), the Singapore Ministry of Education (Grant No. MOE2016-T3-1-006), and the China Scholarship Council (Grant No. 201708440254).

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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