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Substrate-Induced Resonant Magnetoelectric Effects for Dielectric Nanoparticles

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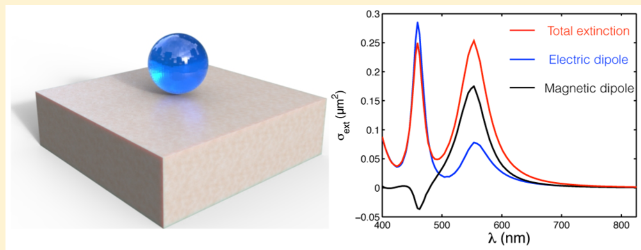
Supporting Information

ABSTRACT: We study, both numerically and analytically, the effect of a substrate on the optical properties of high-refractive-index dielectric nanoparticles. We demonstrate that the optical response of subwavelength nanoparticles can be effectively modeled in the dipole approximation when the dielectric particle is replaced by a pair of point electric and magnetic dipoles with modified polarizabilities. Based on our model, we reveal the existence of the substrate-induced bianisotropic interaction (magnetoelectric coupling) between electric and magnetic dipole modes, which is strongly enhanced near the corresponding dipole resonances. This results in a specific feature of the radiation pattern that can be employed to identify the effective bianisotropic interaction experimentally.



KEYWORDS: *substrate-induced bianisotropy, magnetoelectric coupling, multipole expansion, dyadic Green's function*

The study of light scattering by subwavelength particles is one of the fundamental problems of electrodynamics, which was analytically solved more than 100 years ago.[1] The exact solution is known as the Mie solution, and it was successfully employed for the explanation of various phenomena including the colors of the sky, resonant light scattering and absorption by nanoparticles, photonic jets, as well as optical trapping and optical forces acting on nanoparticles.[2] Recently, this theory was also used to confirm that 50−100 nm silicon nanoparticles scatter light predominantly as magnetic dipoles in the visible range.[3−5] Moreover, recently, a new laser printing method was developed for fabrication of spherical Si nanoparticles with controllable crystallographic phases,[6] allowing for fine-tuning of optical resonances in the visible range.

However, unlike an idealized problem of the Mie scattering in a free space, in many experiments, nanoparticles are placed on a substrate which, in general, might modify substantially their optical response. Thus, several important questions arise: (i) what is the influence of a substrate on the scattering of light by nanoparticles, and (ii) what kind of novel effects are expected in the presence of a substrate? These kinds of problems are significantly important for many applications, including optical nanoantennas, single-molecule spectroscopy, modification of a spontaneous decay rate by nanoparticles, cavity quantum electrodynamics, a design of a nanophotonics circuit, surface-enhanced Raman effects, and many others.

One of the first approaches to this problem was developed by Sommerfeld[7] for a radiating dipole oriented vertically and placed near a planar and lossy surface, which led him eventually to the discovery of surface waves. Since an exact solution to this problem is unavailable, many approximate methods to study the light scattering by spherical particles on a substrate were suggested, including the transfer matrix method and multipole expansion.[8,9] One of the widely used methods is the dyadic Green's function approach for an electric dipole,[10] which was successfully applied to describe the Purcell enhancement factor by fluorescent molecules placed on metallic substrates.[11] Importantly, the problem of light scattering by high-index dielectric nanoparticles on a substrate was considered experimentally only recently,[12−15] and most of the analysis was based on numerical methods.[16−18]

In this paper, we propose a novel approach to analyze the scattering of light by high-index nanoparticles placed on a substrate. It is known that in free space such nanoparticles support both electric and magnetic optically induced resonances in the visible range.[3−5] We assume that the nanoparticle is made of crystalline silicon (dielectric permittivity is taken from ref 19), and it is placed on different substrates such as glass, silicon, or metal. We reveal several novel substrate-induced magnetoelectric phenomena, including the magnetoelectric coupling and resonant bianisotropic interaction of the electric and magnetic dipoles. We extend the point-dipole approximation based on the dyadic Green's function approach[10] by taking into account both electric and magnetic dipole modes of the nanoparticle placed on a substrate. Importantly, in this approach, we replace a silicon nanoparticle with a pair of electric and magnetic dipoles located at the center of the nanoparticle with the substrate-modified polarizabilities. We demonstrate that such dipoles allow us to keep to a

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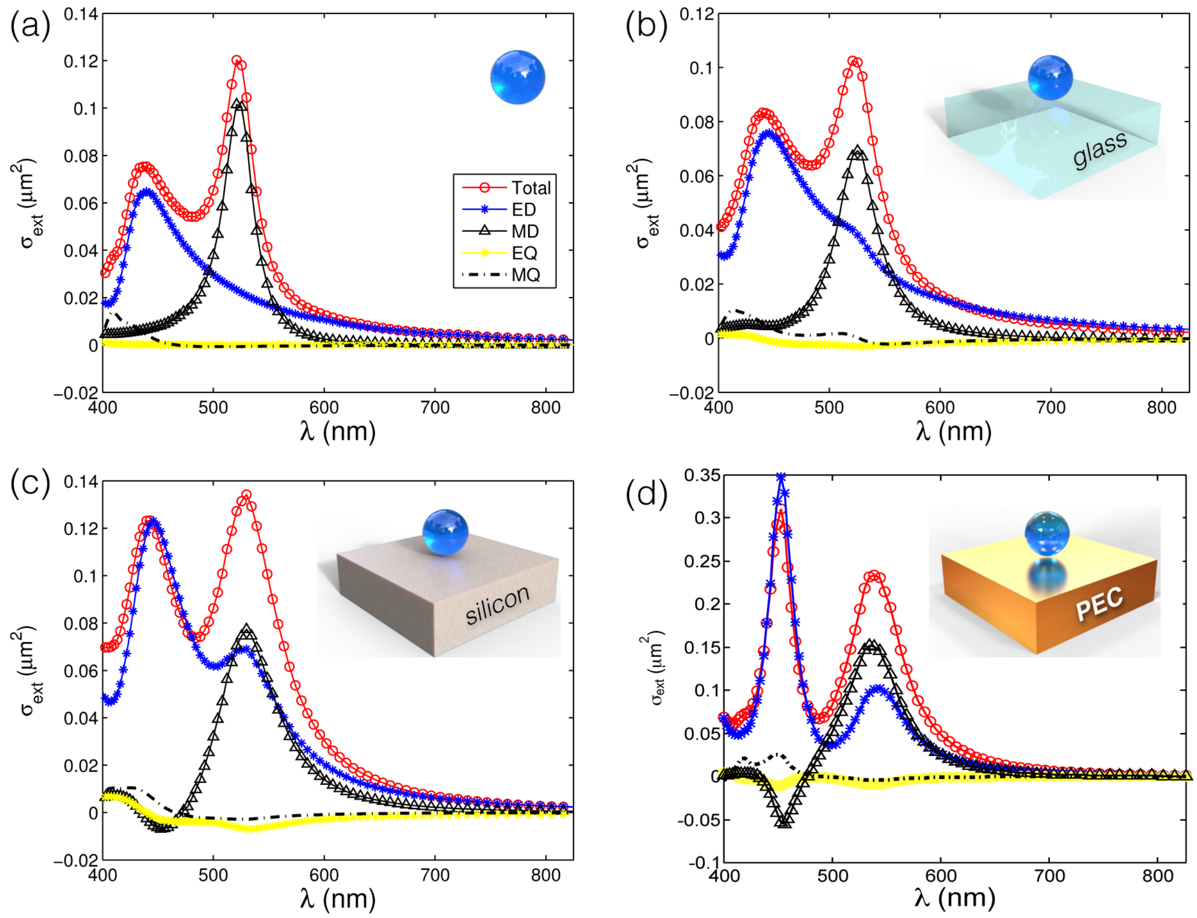

Figure 1. Numerical multipole decomposition of the extinction cross section of a Si spherical nanoparticle with a radius of R = 65 nm for (a) free space, and for the cases when the nanoparticle is placed on (b) a glass substrate with n_{sub} = 1.45. (c) Si substrate and (d) PEC substrate. A particle on a substrate is irradiated by a normally incident linear-polarized plane wave. The multipole expansion is realized with respect to the center of the nanoparticle.

minimum the number of leading terms contributing to the scattering and extinction, in contrast to other approaches where the central point is chosen to be at the substrate interface.[14,20,21]

We believe that our results are important for advanced manipulation of light scattering by nanoparticles in various applications, such as optical nanoantennas and all-dielectric metasurfaces, where the effects of the substrate are expected to be crucially important.

■ RESULTS AND DISCUSSION

Numerical Approach. We start our analysis from direct numerical simulations of the light extinction cross sections of a spherical silicon nanoparticle placed on different substrates. In order to obtain the important information about the contribution of the nanoparticle's multipole moments into the extinction spectra, we employ the so-called decomposed discrete-dipole approximation. This method combines the ordinary discrete-dipole approximation[22,23] with the calculations of the Cartesian multipole moments of electric dipole systems,[24] and details can be found elsewhere.[25] Below, we consider monochromatic fields with the time dependence of $\exp(-i\omega t)$, where ω is the angular frequency.

Figure 1a−c shows the total extinction spectrum, as well as partial contributions from various nanoparticle Cartesian multipoles, calculated for a silicon (Si) nanoparticle located in free space and on top of different substrates; the notations are ED, electric dipole; MD, magnetic dipole; EQ, electric quadrupole; and MQ, magnetic quadrupole. These results clearly demonstrate that, in addition to the primary electric dipole and magnetic dipole resonances observed in free space (Figure 1a), there appear additional cross-coupled substrate-induced bianisotropic resonant contributions. What is more important here is that the substrate-induced contribution can be either *positive*, as a contribution to the electric response at the magnetic dipole resonance (λ = 535 nm), or *negative*, as a contribution to the magnetic response at the electric dipole resonance (λ = 450 nm) (see Figure 1c). These effects manifest themselves stronger by increasing the reflectivity of a substrate, and they reach their maxima for a perfect electric conductor (PEC) substrate (Figure 1d). To identify these substrate-induced bianisotropic contributions, below, we develop a novel theoretical approach that can be termed as the generalized point-dipole approximation.

Note that in Figure 1 the multipole expansion is realized with respect to the center of the nanoparticle. In Figure S1 in the Supporting Information, we present the multipoles' contribution to both cross sections for Si nanoparticles with a radius of R = 65 nm on the PEC substrate, expanded around the nanoparticle center and around a point of the PEC interface. These results demonstrate that in the case of the origin of the multipole expansion at the center of the nanoparticle only two

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leading terms (electric and magnetic dipoles) are sufficient to reconstruct the full extinction and scattering cross sections. Thus, it allows us in the following to replace a dielectric nanoparticle over the substrate by a pair of point electric and magnetic dipoles *located at the center* of the nanoparticle with the substrate-modified polarizabilities, which can be calculated based on the dyadic Green's function method.

Analytical Approach. As follows from the discussed above, the scattering of light by subwavelength particles can be analyzed by using the dipole approximation with the effective polarizabilities

$$\mathbf{p} = \epsilon_0\alpha_E\mathbf{E}_{\text{total}}, \quad \mathbf{m} = \alpha_H\mathbf{H}_{\text{total}} \tag{1}$$

where ϵ_0 is the vacuum permittivity, $\mathbf{p}$ and $\mathbf{m}$ are effective electric and magnetic dipole moments, $\mathbf{E}_{\text{total}}$ and $\mathbf{H}_{\text{total}}$ are total electric and magnetic fields acting on the particle, and $\alpha_{E,H}$ are effective polarizabilities expressed through Mie's dipole scattering coefficients a_1 and b_1, $\alpha_E = 6i\pi a_1/k^3$ and $\alpha_H = 6i\pi b_1/k^3$, where k is a wavenumber.[2,3]

In the presence of a substrate, the total field in eq 1 can be presented in the form

$$\mathbf{E}_{\text{total}} = \mathbf{E}_0 + \mathbf{E}^{\mathbf{p}}_{\text{ref}} + \mathbf{E}^{\mathbf{m}}_{\text{ref}}$$
$$\mathbf{H}_{\text{total}} = \mathbf{H}_0 + \mathbf{H}^{\mathbf{p}}_{\text{ref}} + \mathbf{H}^{\mathbf{m}}_{\text{ref}} \tag{2}$$

where $\mathbf{E}_0$ and $\mathbf{H}_0$ are superpositions of incident and reflected electric and magnetic fields, $\mathbf{E}^{\mathbf{p},\mathbf{m}}_{\text{ref}}$ and $\mathbf{H}^{\mathbf{p},\mathbf{m}}_{\text{ref}}$ are reflected electric and magnetic fields of the corresponding dipoles that can be expressed by using the reflection dyadic Green's function calculated at the point of excitation

$$\mathbf{E}^{\mathbf{p}}_{\text{ref}} = \hat{G}^{\text{ref}}_{\text{ee}}(\mathbf{r}_0, \mathbf{r}_0)\mathbf{p} = \frac{\mu k^2}{\epsilon_0}\hat{G}^{E}_{\text{ref}}(\mathbf{r}_0, \mathbf{r}_0)\mathbf{p}$$
$$\mathbf{E}^{\mathbf{m}}_{\text{ref}} = \hat{G}^{\text{ref}}_{\text{em}}(\mathbf{r}_0, \mathbf{r}_0)\mathbf{m} = i\omega\mu\mu_0[\nabla \times \hat{G}^{H}_{\text{ref}}(\mathbf{r}_0, \mathbf{r}_0)]\mathbf{m}$$
$$\mathbf{H}^{\mathbf{p}}_{\text{ref}} = \hat{G}^{\text{ref}}_{\text{me}}(\mathbf{r}_0, \mathbf{r}_0)\mathbf{p} = -i\omega[\nabla \times \hat{G}^{E}_{\text{ref}}(\mathbf{r}_0, \mathbf{r}_0)]\mathbf{p}$$
$$\mathbf{H}^{\mathbf{m}}_{\text{ref}} = \hat{G}^{\text{ref}}_{\text{mm}}(\mathbf{r}_0, \mathbf{r}_0)\mathbf{m} = \epsilon\mu k^2\hat{G}^{H}_{\text{ref}}(\mathbf{r}_0, \mathbf{r}_0)\mathbf{m}$$

where ϵ and μ are relative permittivity and permeability of upper superstate, ω is a frequency of light, and $\mathbf{r}_0 = \{x_0, y_0, z_0\}$ is the position of the dipole. The substrate interface lies in $x-y$ plane wit $z = 0$. We can now solve the system (2) with respect to the electric and magnetic dipoles (1)

$$\mathbf{p} = \epsilon_0\hat{\alpha}_{EE}\mathbf{E}_0 + \hat{\alpha}_{EM}\mathbf{H}_0 \tag{3}$$

$$\mathbf{m} = \hat{\alpha}_{MM}\mathbf{H}_0 + \hat{\alpha}_{ME}\mathbf{E}_0 \tag{4}$$

where

$$\hat{\alpha}_{EE} = \hat{A}^{-1}\alpha_E, \quad \hat{\alpha}_{MM} = \hat{B}^{-1}\alpha_H \tag{5}$$

$$\hat{\alpha}_{EM} = \hat{A}^{-1}\epsilon_0\alpha_E\hat{G}^{\text{ref}}_{\text{em}}(\hat{I} - \alpha_H\hat{G}^{\text{ref}}_{\text{mm}})^{-1}\alpha_H \tag{6}$$

$$\hat{\alpha}_{ME} = \hat{B}^{-1}\alpha_H\hat{G}^{\text{ref}}_{\text{me}}(\hat{I} - \epsilon_0\alpha_E\hat{G}^{\text{ref}}_{\text{ee}})^{-1}\epsilon_0\alpha_E \tag{7}$$

with

$$\hat{A} = \hat{I} - \epsilon_0\alpha_E\hat{G}^{\text{ref}}_{\text{ee}} - \epsilon_0\alpha_E\hat{G}^{\text{ref}}_{\text{em}}(\hat{I} - \alpha_H\hat{G}^{\text{ref}}_{\text{mm}})^{-1}\alpha_H\hat{G}^{\text{ref}}_{\text{me}}$$
$$\hat{B} = \hat{I} - \alpha_H\hat{G}^{\text{ref}}_{\text{mm}} - \alpha_H\hat{G}^{\text{ref}}_{\text{me}}(\hat{I} - \epsilon_0\alpha_E\hat{G}^{\text{ref}}_{\text{ee}})^{-1}\epsilon_0\alpha_E\hat{G}^{\text{ref}}_{\text{em}}$$

and $\hat{I}$ is (3×3) unitary tensor.

One of the interesting cases is a PEC substrate, which can be solved analytically. Being an ideal mirror, PEC should exhibit the strongest substrate-induced bianisotropic effects, but at the same time, it simplifies substantially the analytical approach, reducing the problem, basically, to the method of images. For the normal incidence of the x-polarized light at the PEC substrate, the excitation fields become

$$\mathbf{E}_0 = \{-2i\sin(kz_0), 0, 0\} \tag{8}$$

$$\mathbf{H}_0 = \frac{1}{\eta}\{0, -2\cos(kz_0), 0\} \tag{9}$$

where μ_0 is the vacuum permittivity, and $\eta = (\mu_0/\epsilon_0)^{1/2}$ is the free-space wave impedance. The dyadic Green's functions for the PEC substrate have the following form

$$\hat{G}^{E}_{\text{ref}}(\mathbf{r}_0, \mathbf{r}_0) = -\hat{G}^{H}_{\text{ref}}(\mathbf{r}_0, \mathbf{r}_0) = \begin{pmatrix} A & 0 & 0 \\ 0 & A & 0 \\ 0 & 0 & B \end{pmatrix} \tag{10}$$

$$\nabla \times \hat{G}^{E}_{\text{ref}}(\mathbf{r}_0, \mathbf{r}_0) = -\nabla \times \hat{G}^{H}_{\text{ref}}(\mathbf{r}_0, \mathbf{r}_0) = \begin{pmatrix} 0 & C & 0 \\ -C & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{11}$$

with

$$A = \frac{1 - 2kz_0(i + 2kz_0)}{32k^2\pi z_0^3}e^{2ikz_0}$$
$$B = -\frac{2ikz_0 - 1}{32\pi z_0^2 k^2}e^{2ikz_0}$$
$$C = \frac{2ikz_0 - 1}{16\pi z_0^2}e^{2ikz_0}$$

It is important to notice that, due to a specific structure of reflected dyadic Green's function tensor (see eqs 10 and 11 and similar expressions in Supporting Information), for an arbitrary substrate, the cross-coupling polarizability terms are equal to each other up to the normalization factor μ_0

$$\hat{\alpha}_{EM} = \mu_0\hat{\alpha}_{ME} = \begin{pmatrix} 0 & D & 0 \\ -D & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \tag{12}$$

where the value D depends on the material properties of a substrate (see Supporting Information). It demonstrates that for a particle placed on a substrate the induced bianisotropic coupling between the electric and magnetic modes is in-phase. Another observation is that only in-plane components of the exciting fields will contribute to the bianisotropic response, which will be discussed in detail below.

The total extinction cross section can now be written in the form[3]

$$\sigma_{\text{ext}} = \sigma^{\mathbf{m}}_{\text{ext}} + \sigma^{\mathbf{p}}_{\text{ext}} \tag{13}$$

with

$$\sigma^{\mathbf{m}}_{\text{ext}} = \frac{\omega\mu_0}{2P_{\text{in}}}\text{Im}(\mathbf{H}^*_0\cdot\mathbf{m}); \quad \sigma^{\mathbf{p}}_{\text{ext}} = \frac{\omega}{2P_{\text{in}}}\text{Im}(\mathbf{E}^*_0\cdot\mathbf{p}) \tag{14}$$

where $\sigma^{\mathbf{m}}_{\text{ext}}$ and $\sigma^{\mathbf{p}}_{\text{ext}}$ are the contributions of the magnetic dipole and electric dipoles to the total extinction cross section,

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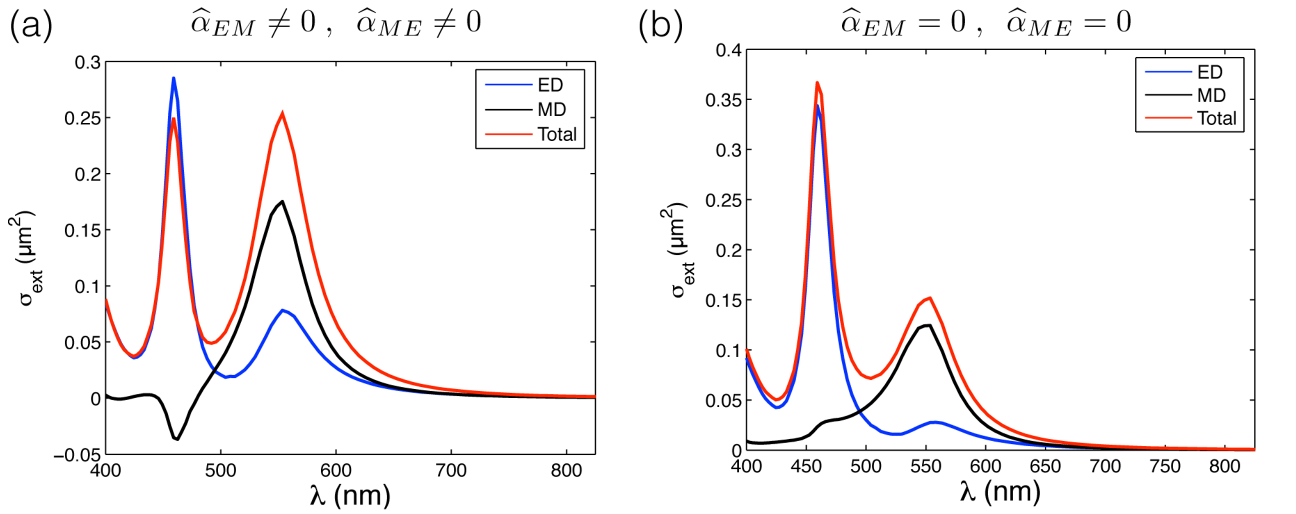

Figure 2. Extinction cross section for Si nanoparticle with a radius of $R = 65$ nm calculated in the point electromagnetic dipole approximation for the PEC substrate (a) with and (b) without cross-coupling terms (6 and 7), demonstrating the role of substrate-induced modes overlapping (hybridization) with negative interference contribution caused by bianisotropy.

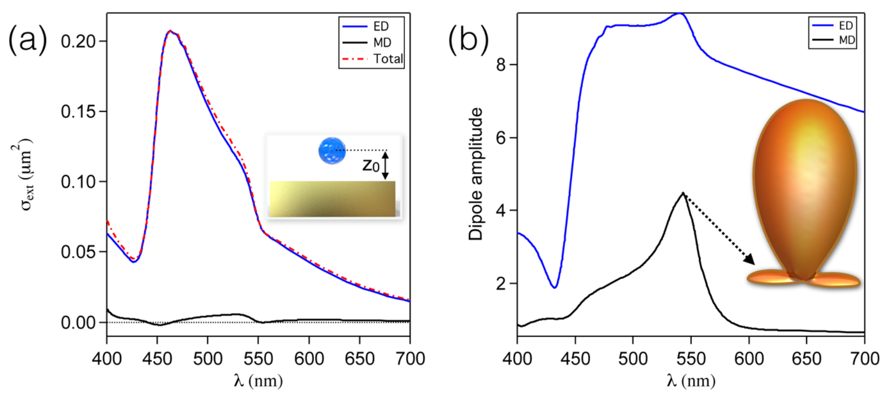

Figure 3. (a) Extinction cross section and (b) amplitudes of the electric and magnetic dipole modes for a Si nanoparticle with $R = 65$ nm placed on the PEC substrate at the distance $z_0 = 138.175$ nm at the node of the incident magnetic field $H_0 = 0$ for the wavelength $\lambda = 553.5$ nm (8). Inset shows the angular distribution of the scattering field at the magnetic resonance with the characteristic side lobes due to the induced bianistropy effect.

respectively, and P_{in} is the radiation flux of the incident wave.[24,25] All details of the point-dipole approximation in the presence of the substrate can be found in Supporting Information.

Resonant Magnetoelectric Coupling. The analytical expressions 3 and 4 describe a number of interesting effects that allow us to understand deeper the substrate-induced dipole contributions into the extinction cross section. First, the induced electric and magnetic dipoles can now be excited by both electric and magnetic fields *independently*, which results in a substrate-induced magnetoelectric coupling, also known as *bianisotropy response*.[26] Moreover, at the corresponding resonances, the interaction between induced dipoles is significantly enhanced, leading to *resonant cross-coupling interaction*. In Figure 2a, we plot the total extinction as well as partial contributions from the electric and magnetic dipoles, calculated with the point-dipole approximation for the PEC substrate by using eqs 1−14. Figure 2a is in a good qualitative agreement with the results presented in Figure 1d, which justifies the validity of our method. From Figure 2a, one can clearly see that, in addition to the primary dipole resonances which exist without any substrate, there appear to be cross-coupled substrate-induced resonant contributions for both electric and magnetic responses. In Figure 2a,b, we show a comparison between the full solutions (Figure 2a) and a reduced solution where the bianisotropy terms (eqs 3 and 4) that are proportional to $\hat{\alpha}_{EM}$ and $\hat{\alpha}_{ME}$ vanish (see Figure 2b). In the latter case, there is no negative contribution to the extinction cross section at the MD response. Thus, we come to the conclusion that an external magnetic field extorts a negative work on the magnetic dipole at the electric resonance. In other words, a part of the energy extracted resonantly from the incident wave by the electric dipole is returned back by the resonantly excited magnetic dipole mode. Physically, different-sign (positive or negative) ED and MD contributions into the total extinction cross section correspond to a *destructive interference* between the fields solely scattered by these dipole moments of nanoparticles.

Since the plane wave impinged on the substrate forms a standing wave (8 and 9), it becomes possible to separate clearly the bianisotropy effect by choosing a proper elevation of the nanoparticle above the substrate. Indeed, it is possible to find the conditions when the particle with the induced magnetic dipole resonance will be at a node of the incident magnetic field $\mathbf{H}_0 = 0$. It implies that the magnetic dipole contribution to the extinction should vanish, as well (see eq 14), namely, $\sigma_{ext}^{m} = 0$.

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ACS Photonics 2015, 2, 1423−1428

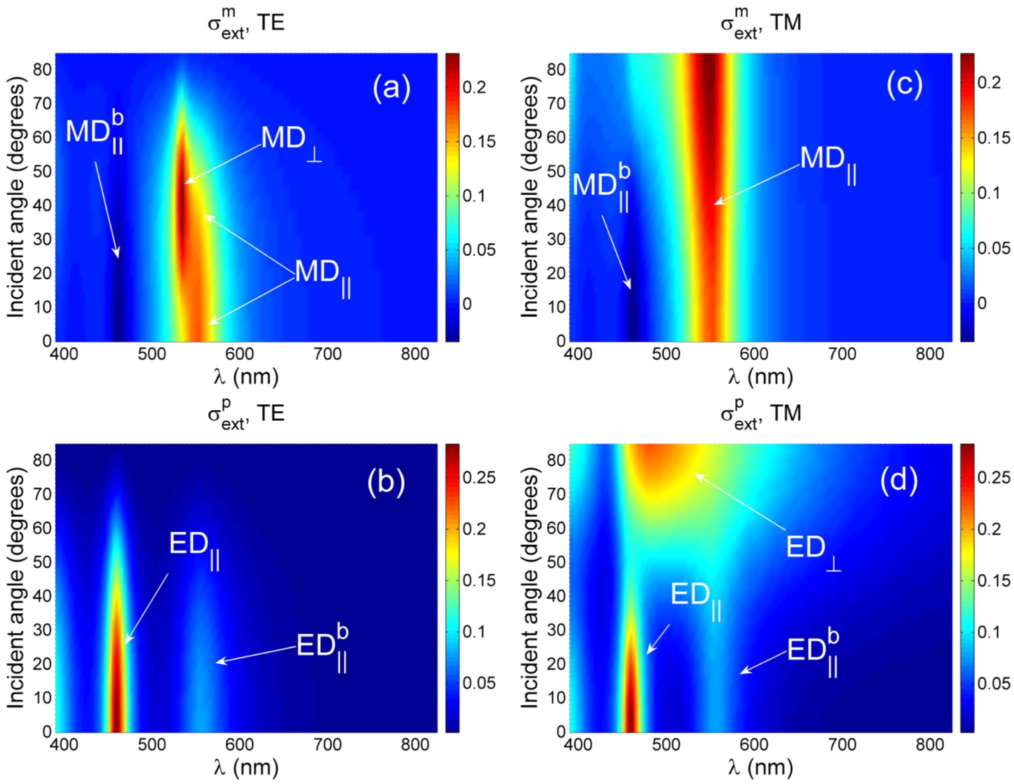

Figure 4. (a,c) Magnetic dipole σ^{m}_{ext} and (b,d) electric dipole σ^{p}_{ext} contributions (μm^2) to the total extinction cross section (eq 13) calculated for a Si nanoparticle (R = 65 nm) located on the PEC substrate as a function of the incident angle and wavelength of light. (a,b) TE-polarized and (c,d) TM-polarized incident light. Arrows indicate the resonant contributions of the corresponding dipole moments: $MD_{\parallel}$ ($ED_{\parallel}$) and $MD_{\perp}$ ($ED_{\perp}$) are the in-plane magnetic (electric) dipole resonance and out-of-plane magnetic (electric) dipole resonance excited by external waves; $MD^{b}_{\parallel}$ ($ED^{b}_{\parallel}$) is the magnetic (electric) dipole resonance excited due to the resonant bianisotropy effect.

Moreover, without bianisotropy, there will be no magnetic dipole response at all (according to eq 1, **m** should vanish). However, according to our solutions 3 and 4, we still expect a strong magnetic response due to the substrate-induced magnetoelectric coupling. Figure 3a demonstrates that under this condition the dominant contribution to the total extinction cross section for a Si nanoparticle is from the electric dipole response, while the magnetic contribution is quite weak, and it even vanishes at the magnetic dipole resonance. At the same time, the magnetic dipole mode is still resonantly excited at its resonant wavelength due to bianisotropy (see Figure 3b). This also results in a peculiar radiation pattern, with the induced side lobes corresponding to the magnetic dipole excitation (see the inset in Figure 3b), an effect that can be observed experimentally.

Oblique Incidence. By using the analytical approach for the point-dipole approximation, the ED and MD contributions to the extinction cross sections of the Si nanoparticle can be analyzed for the case of the oblique incidence as functions of the incident angle and wavelength; see the results in Figure 4 for both polarizations. Due to the resonant magnetoelectric coupling, the bianisotropic components of the magnetic and electric dipoles $MD^{b}_{\parallel}$ and $ED^{b}_{\parallel}$ correspond only to in-plane field components of an incident light wave. Thus, their incident angular distributions replicate the distributions of the corresponding in-plane resonances, namely, $MD^{b}_{\parallel} \leftrightarrow ED_{\parallel}$ and $ED^{b}_{\parallel} \leftrightarrow MD_{\parallel}$. No bianisotropy responses correspond to the out-of-plane $MD_{\perp}$ and $ED_{\perp}$ resonances in Figure 4. In contrast to the broad out-of-plane $ED_{\perp}$ resonance (see Figure 4d) excited at large incident angles, the out-of-plane $MD_{\perp}$ resonance (see Figure 4a) is spectrally narrow with a maximum at the incident angle of ~40°. We notice that the magnetic dipole resonances $MD^{b}_{\parallel}$ excited due to the bianisotropic effect have similar incident angle dependence for both polarizations. The negative contributions of $MD^{b}_{\parallel}$ in σ^{m}_{ext} (see Figure 4a,c) decrease with an increase of the incident angle due to the angular dependence of the in-plane component $\mathbf{E}_{0\parallel}$ of the external electric field $\mathbf{E}_0$: $\mathbf{E}_0^{TE} \sim \sin(kz_0 \cos\beta)$ and $\mathbf{E}_{0\parallel}^{TM} \sim \cos(\beta)\sin(kz_0 \cos\beta)$, where β is the angle of incidence. Indeed, according to eq 4, the bianisotropic part of MD is proportional to the in-plane component of the exciting electric field. In contrast to the case of $MD^{b}_{\parallel}$, the bianisotropic electric dipole resonances $ED^{b}_{\parallel}$ depend differently on the incident angle for two polarizations. In the case of TE polarization (TM polarization), the contributions of $ED^{b}_{\parallel}$ into the extinction cross section are decreasing (increasing) with a growth of the incident angle because of the incident angular dependence of the magnetic field in-plane component, $\mathbf{H}_{0\parallel}^{TE} \sim \cos(\beta)\sin(kz_0 \cos\beta)$ and $\mathbf{H}_0^{TM} \sim \cos(kz_0 \cos\beta)$, which determines the bianisotropic part of the ED contribution (see eq 3).

CONCLUSIONS

We have studied the scattering of light by a subwavelength dielectric nanoparticle placed on a substrate. We have demonstrated that, by performing the multipole expansion around the center of the particle, it is sufficient to keep electric and magnetic dipoles to fully recover the extinction and scattering cross sections. This allows us to employ the dyadic Green's function method and point-dipole approximation to study analytically the optical response of dielectric nano-

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particles placed on substrates. Due to the coexistence of both electric and magnetic dipole responses, we have predicted and demonstrated several novel substrate-induced magnetoelectric effects, including the renormalization of the resonance frequencies, mode overlap, magnetoelectric coupling, and resonant bianisotropy. Based on our analysis, we have suggested how different effects can be observed experimentally in the scattering measurements. The understanding of a role of a substrate is crucially important for many applications, including sensing, optical nanoantennas, and optical trapping. We believe that our findings will provide rigorous and complete grounds for solving many such problems, also allowing for quantitative evaluation of the substrate-induced effects.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsphotonics.5b00117.

Direct numerical simulations showing where the effective dipoles should be located; generalized analytical approach for a substrate made of an arbitrary material based on the dyadic Green's function approach (PDF)

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Notes

The authors declare no competing financial interest.

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