Enhancement of artificial magnetism via resonant bianisotropy

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All-dielectric “magnetic light” nanophotonics based on high refractive index nanoparticles allows controlling magnetic component of light at nanoscale without having high dissipative losses. The artificial magnetic optical response of such nanoparticles originates from circular displacement currents excited inside those structures and strongly depends on geometry and dispersion of optical materials. Here an approach for enhancing of magnetic response via resonant bianisotropy effect is proposed and analyzed. The key mechanism of enhancement is based on electric-magnetic interaction between two electrically and magnetically resonant nanoparticles of all-dielectric dimer. It was shown that proper geometrical arrangement of the dimer in respect to the incident illumination direction allows flexible control over all vectorial components of the magnetic moment, tailoring the latter in the dynamical range of 100% and delivering enhancement up to 36% relative to performances of standalone spherical particles. The proposed approach provides pathways for designs of all-dielectric metamaterials and metasurfaces with strong magnetic responses.

Intrinsic magnetic polarizabilities of natural materials have strong frequency dependence with the fundamental cut-off in GHz range, originating from relatively low spin and orbital susceptibilities1. Recently, polarization currents in subwavelength structured loops, organized in ordered arrays, became sources of high-frequency artificial magnetism2,3. Nanostructured noble metals, supporting localized plasmon resonances could serve as building blocks for metamaterials with artificial magnetic polarizability4. However, inherent material losses set severe limitations on performances of such structures5. Another approach for obtaining magnetic optical response is to employ circular displacement currents in high-index dielectric nanoparticles6,7. This is the essence of so-called all-dielectric nanophotonics, which opened the way to control magnetic component of light at nanoscale without high dissipation, inherent for metallic nanostructures8–14. The “magnetic light” concept found use in various applications, such as nanoantennas14, quantum interface for NV-centers16, photonic topological insulators17, broadband perfect reflectors18, waveguides19, cloaking20, harmonics generation21, wave-front engineering, and dispersion control22, fluorescence enhancement23, and many more.

Magnetic response of a dielectric particle strongly depends on its refractive index, shape, and external environment. The eigen frequencies of electric and magnetic resonances could span the entire visible range. However, the magnitude of those multipole moments is limited by dispersion properties of optical materials.

Here an approach for tailoring magnetic response of dielectric nanoparticles via resonant bianisotropy is proposed. Microscopically, bianisotropy is the effect of magneto-electric coupling, where electric polarization induces magnetic and vice versa. The signature of this effect appears in the constitutive relations, e.g. the dependence of electrical induction also on magnetic field and magnetic induction on electric field23,24. Bianisotropy is used for achieving high values of effective polarizabilities in metamaterials25, unique properties of metasurfaces26,27, and designed directivity of nanoantennas28. Previously, it was shown that the interaction of dielectric nanoparticles with substrates may increase induced magnetic moment due to the effect of non-resonant bianisotropy29,30. Resonant interaction between a nanoparticle and a surface was analysed in ref. 31. The approach, proposed here, is based on electric-magnetic interaction between two resonant nanoparticles of a dimer (see Fig. 1). The nanoparticles were designed in the following way: the electric dipolar resonance of the bigger sphere spectrally overlaps with magnetic response of the smaller one. In this case, the effect of resonant bianisotropy is...
achieved: the resonant electric moment of the bigger nanoparticle induces the additional magnetic moment in the smaller one, tailoring its overall response. It is worth noting, that non-resonant overlapping regime was previously considered in ref. 32.

The manuscript is organized as follows: first, the optical properties of isolated spherical particles are briefly discussed in the context of the resonance tuning. Next, the analytical coupled dipoles formulation of the problem is developed and verified with numerical modeling. The expression describing the magnetic moment of the nanoparticle, with account for the bianisotropy, is derived. Finally, it is shown that proper geometrical arrangement of the dimer in respect to the incident illumination direction allows achieving additional vectorial component of magnetic polarization.

Results and Discussions

Coupled dipoles theory and numerical modelling. In order to obtain the dimer design, properties of isolated components will be briefly discussed. First, isolated silicon sphere of radius $R_2 = 52 \text{ nm}$, having magnetic dipolar resonance at wavelength $477 \text{ nm}$, i.e. in the visible range is considered. The material dispersion of crystalline silicone (c-Si) is taken from ref. 33. The scattering cross-section of the nanoparticle has been calculated using FDFD (Finite-Difference Frequency Domain) simulations in CST Microwave Studio, and corresponding results as the function of wavelength are presented in Fig. 2(a) (blue curve). Electric dipolar resonance of 70-nm radius particle overlaps with the magnetic dipolar resonance of 52-nm sphere at $\lambda \approx 480 \text{ nm}$.

Figure 1. The geometry of the bianisotropic all-dielectric dimer. The system consists of two dielectric nanoparticles separated by a distance $r_{12}$. The sizes of nanoparticles are $R_1$ (bigger nanoparticle) and $R_2$ (smaller nanoparticle). Electric dipole resonance of the big sphere overlaps with magnetic dipole resonance of the small one. The dimer is excited by a $y$-polarized plane wave propagating along $x$-axis.

Figure 2. Optical properties of single silicon nanoparticles with radii $R_1 = 70 \text{ nm}$ (red curves) and $R_2 = 52 \text{ nm}$ (blue curves). (a) The scattering cross-section spectra, normalized to geometric cross-section ($\pi r^2$). (b) Dispersion of particles polarizabilities. The curves correspond to analytical calculations, red circles and blue squares represent numerical results. Electric dipolar resonance of 70-nm radius particle overlaps with the magnetic dipolar resonance of 52-nm sphere at $\lambda \approx 480 \text{ nm}$. 

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where 52-nm sphere exhibits resonant magnetic response (Fig. 2a, red curve). Resonance overlapping effect can be achieved in a single cylindrical or elliptical particle (e.g. ref. 34). The electric and magnetic polarizabilities which are associated with induced electric and magnetic moments have been also calculated (Fig. 2b). Results of numerical simulations (dots) are in good agreement with analytical Mie theory (curves) verifying the validity of the numerical tool. Values of those electric and magnetic moments will be subsequently used in the analytical model, based on discrete dipole approximation.

Next, the scattering of a plane wave on all-dielectric dimer will be analyzed. The electromagnetic scattering problem could be solved by employing Coupled Electric and Magnetic Dipole Approximation (CEMDA). In this method, complex nanostructures are represented by converging series of point electric and magnetic dipoles, while the problem of two spheres could be approximated by only two. This approach is particularly accurate, if the gap between the spheres is bigger than their radii, however it could be still applicable for the case of small separation distances, as it was shown in ref. 29. The goal of the subsequent analytical analysis is obtaining a simple formulation, underlining the interference phenomena, affecting the magnetic dipole moment of the smaller particle and revealing the bianisotropic nature of the interaction. The impact of higher order modes has been taken into account using the full-wave numerical calculations. Following the CEMDA method, total electromagnetic fields [both electric (E) and magnetic (H)] were decomposed into incident and scattered components:

\[ E(r_j) = E_0(r_j) + E_s(r_j), \]
\[ H(r_j) = H_0(r_j) + H_s(r_j), \]

where indices \( i, j = 1, 2 \) (\( i \neq j \)) denote the first (bigger) and the second (smaller) nanoparticle, respectively, \( E_0 \) and \( H_0 \) are electric and magnetic fields of the incident plane wave, \( E \) and \( H \) are the total electric and magnetic fields, \( E_s \) and \( H_s \) are the electric and magnetic fields scattered by the nanoparticle with index \( j \), \( r \) is a radius-vector of \( j \)-nanoparticle's center. Both particles are polarized by the incident field, as well as by the scattered one:

\[ p_i = \alpha_i^E E(r_i), \quad m_i = \alpha_i^M H(r_i), \]

where \( \alpha_i^E \) and \( \alpha_i^M \) are the electric and magnetic polarizabilities of a single particle “i”:

\[ \alpha_i^E = \frac{6\pi\epsilon_0\epsilon_b}{k_0^2} a_i, \quad \alpha_i^M = \frac{6\pi}{k_0^2} b_i, \]

and \( \epsilon_b \) is the dielectric constant of surrounding medium, \( \epsilon_0 \) is the vacuum permittivity, \( j \) stands for the imaginary unit. The coefficients \( a_i \) and \( b_i \) are the first order Mie scattering coefficients, and are given by: \( a_i(\lambda) = [A - B]/[C - D], b_i(\lambda) = [Bn^{-2} - A]/[Dn^{-2} - C], \) where the following notation is introduced:

\[
A = \left[ \frac{\cos(\rho n)}{\rho n} + \frac{\sin(\rho n)}{(\rho n)^2} \right] \left[ \frac{\sin(\rho)}{(\rho n)^2} - \frac{\cos(\rho)}{\rho^2} \right], \\
B = n^2 \left[ \frac{\cos(\rho)}{\rho} + \frac{\sin(\rho)}{\rho^2} \right] \left[ \frac{\sin(\rho)}{(\rho n)^2} - \frac{\cos(\rho)}{\rho^2} \right], \\
C = - \left[ \frac{\cos(\rho n)}{\rho n} + \frac{\sin(\rho n)}{(\rho n)^2} \right] \left[ \frac{\sin(\rho)}{(\rho n)^2} - \frac{1}{\rho^2} \right] e^{j\rho}, \\
D = n^3 \left[ \frac{e^{j\rho}(j + \rho j^2)}{\rho^2} \right] \left[ \frac{\sin(\rho)}{(\rho n)^2} - \frac{\cos(\rho)}{\rho n} \right],
\]

where \( n = \sqrt{\epsilon_b/\epsilon_0}, \rho = k_0 R_i, R_i \) - radius of the sphere “i”, \( \epsilon \) - the permittivity of the material (silicon, here), \( k_0 = 2\pi/\lambda \) - the wave number in the surrounding medium, and \( \lambda \) is a free-space wavelength.

The values of single particle polarizabilities, calculated using Eqs. (3) and (4), are in a perfect agreement with our numerical calculations [see Fig. 2(b)]. Thus, the scattered fields of the dipoles can be obtained through the Green’s function of a point dipole in a free-space \( G(r, r_j) \equiv G_{ij} \), following the CEMDA:

\[
p_i = \alpha_i^E \left[ E_0(r_i) + \frac{k_0^2}{\epsilon_0} \frac{G_{ij}}{G_{ij}} [g_j \times m_j] \right], \\
m_i = \alpha_i^M \left[ H_0(r_i) + \frac{k_0^2}{\epsilon_0} \frac{G_{ij}}{G_{ij}} [g_j \times p_j] \right],
\]

where \( k_0 \) and \( c \) are the wavenumber and speed of light in vacuum. The Green’s function of a point dipole in a free-space is given by:
where \( \mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j \) is the radius-vector connecting the first dipole (coordinates origin, or the center of the bigger particle) with the second one (center of the smaller particle), \( r_{ij} = |\mathbf{r}_{ij}| \). The tensor \( G_{ij} \) and the vector \( \mathbf{g}_{ij} \) have the following symmetry of indices permutation:

\[
G_{ij} = G_{ji} \quad \text{and} \quad \mathbf{g}_{ij} = -\mathbf{g}_{ji}.
\]

Both nanoparticles, being isolated, have 3-fold degenerated (magnetic and electric) dipolar resonances, oriented along the unit vectors of a Cartesian coordinate system. The excitation of an isolated sphere is solely defined by the polarization of the incident wave – for example, linearly polarized beam will excite only one of the 3 components of the dipolar mode. However, the geometry of the coupled dipoles together with the excitation (not necessarily coinciding with one of the symmetry axis of the system) will break the degeneracy and, as the result, all three vectorial components should be taken into account. The resulting set of equations can be solved analytically by means of the matrix inversion or numerically in the same fashion. In order to verify the validity of the proposed theoretical model, a particular case, where the system of Eqs. (5) has a simple and intuitive solution, was considered. Hereinafter the angles are introduced in accordance with a standard definition of the spherical coordinate system: \( \theta \) is the angle between the vector and the axis \( z \), \( \phi \) is the angle between the vector projection on the \( xy \) plane and \( x \) axis. Arranging the nanoparticles along the \( x \)-axis and exciting the system with plane wave linearly polarized along \( y \)-axis with angles being \( \theta = \pi/2 \) and \( \phi = 0 \), the set of 12 coupled equations was reduced just for 4, since the symmetry considerations allow the moment components to be induced only along \( z \)-axis (magnetic dipole) and \( y \)-axis (electric dipole). This set of coupled equations has particularly simple solution, which agrees well with the numerical calculations. The values of the magnetic moment enhancement, calculated both analytically and numerically as the function of the distance \( r_{12} \) between nanoparticles are shown in Fig. 3. It is clearly seen, that the analytical and numerical models agree well with each other, 15% enhancement of the magnetic moment of the single nanoparticle, as the result of the bianisotropic coupling, can be also observed. For clear understanding of the oscillatory behaviour of magnetic moments, the simplest model, when the polarizabilities \( \alpha_M^E \) and \( \alpha_E^M \) are supposed to be zero, was considered. In this case the simple formula can be obtained, underlining the bianisotropic nature of the effect:

\[
m_2 = \eta H_0 + \gamma E_0, \quad \eta = \frac{\alpha_2^M e^{jKz_1}}{1 - \alpha_1^E \alpha_2^M g_{ij} K_0^2}, \quad \gamma = -j \frac{\varepsilon_0 \alpha_1^E \alpha_2^M g_{ij} K_0^2}{1 - \alpha_1^E \alpha_2^M g_{ij} K_0^2}.
\]  

The qualitative analysis of magnetic moment \( m_2 \) enables observing the clear interference phenomena – the direct excitation of magnetic dipole by the plane wave \( \alpha_2^M e^{jKz_1} \) term in Eq. (7)] and the contribution of the scattered field through the term \( \alpha_1^E \alpha_2^M \). Thus, the oscillation behavior of magnetic moment enhancement is the
result of the interference phenomenon (see Fig. 3). For larger distances the coupling between the particles becomes weaker and converges to the value of the isolated particle. An almost perfect fit of full numerical modeling with the CEMDA method enables to use the latter for analysis of more complex structures without involving full wave simulations. This approach will be subsequently employed.

**Vectorial structure of magnetic moments.** In the subsequent studies the illumination was chosen to propagate along $x$-axis and being polarized along $y$. There are three geometrical parameters, affecting the magnetic moment of the smaller particle: the distance between the spheres’ centres $r_{12} = R_1 + R_2 + D$, and the angles $\theta$ and $\varphi$. The angular dependence of the induced magnetic moment components of the smaller particle for 3 characteristic separation distances $D = 10, 100, 200$, and $300$ nm (gap between the nanoparticles’ surfaces) will be studied next. In the case of noninteracting nanoparticles there is the only one non-zero component of nanoparticle’s magnetic moment $m^z_2$, co-directed with the magnetic field. In the case of bianisotropic coupling there are also $x$- and $y$- non-zero components of the magnetic moment ($m^x_2$, $m^y_2$). Figure 4 shows the three-dimensional angular dependencies of normalized magnetic moment components relative increase ($\alpha^l_2 - m^l_2 H_0$) for all vectorial components $l = x, y, z$ for the different distances $D = 10, 100, 200$, and $300$ nm.
plex structures such as all-dielectric metamaterials and metasurfaces with strong magnetic responses. The proposed approach can find use in designs of more complex structures, e.g., where the magnetic moment components and amplitude values, allowing full on-demand control of the electromagnetic responses at the desired wavelength in the visible range by specifying their radii and materials, a system of two spheres was proposed. While standalone nanoparticles allow obtaining dipolar and high-multipolar resonant responses at the desired wavelength in the visible range by specifying their radii and materials, a system of two coupled nanoparticles possesses more degrees of freedom. Altering the radii of both nanoparticles allows investigating the impact of all the combinations of multipolar coupling effects on the properties of the system. Naturally, the amplitude of the effects is dependent on their mutual displacement, and it decreases at larger distances between the particles, as it was proved both analytically and numerically in the case of dipolar magnetic-electric coupling. Symmetry considerations allow exciting only one dominant induced electric and magnetic moment component when a plane wave is incident upon a single nanoparticle. It was shown, that for the nanoparticle system, altering the spherical angles and distance between nanoparticles allows to excite all vectorial components of magnetic moment simultaneously. The secondary components being up to 50% of the amplitude value of the dominant one. Furthermore, these parameters also define the spectral position of dips and peaks of the dominant electric moment components and amplitude values, allowing full on-demand control of the electromagnetic properties of the system of coupled nanoparticles. The proposed approach can find use in designs of more complex structures such as all-dielectric metamaterials and metasurfaces with strong magnetic responses.

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Author Contributions
P.G., P.B. and A.S. supervised the project. A.K. and K.B. developed theoretical model. K.B. and D.M. performed numerical simulations. All authors contributed to the manuscript preparation.

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