Antiferromagnetism of hybrid metamaterials

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We analyze a metal-dielectric structure composed of a silicon nanoparticle coupled to a stack of split-ring resonators, and reveal the possibility of optically-induced *antiferromagnetic response* of such a hybrid meta-molecule with a staggered pattern of the induced magnetization. We show that a hybrid metamaterial created by a periodic lattice of the meta-molecules supports antiferromagnetic modes with a checker-board pattern and the propagation of spin waves, opening new ways for manipulating artificial antiferromagnetism at high frequencies with low-loss materials.

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Introduction. Metamaterials offer many novel possibilities to manipulate and control the propagation of electromagnetic waves [1, 2]. Typical metamaterials are created by a system of resonant subwavelength elements for which electric and magnetic responses can be varied and modified independently. During last decade there were many efforts to achieve artificial magnetism at high frequencies exploiting the inductive response of normal metals [3]. The idea was successfully implemented in mesh-like metal-dielectric "fishnet" structures [3] and arrays of split-ring resonators [4]. However, in almost all such cases the optically-induced *ferromagnetic* response of the whole structure was utilized, based on the induced magnetization of individual meta-elements [5–8]. The research in this area was stimulated by a possibility to achieve the overall negative magnetic response required for a design of left-handed metamaterials.

A canonical subwavelength artificial magnetic meta-atom is a split-ring resonator (SRR) that consists of an inductive metallic ring with a gap. The basic physical principle behind this design is that a SRR can support a principal eigenmode with a circular current distribution that gives rise to a local magnetic dipole moment [9]. It allows to consider a single SSR as a magnetic counterpart of the conventional Lorentz oscillator model for electric dipoles. The SRR-based design was the first and most frequently used for metamaterials, with unprecedented values of negative magnetic permeability at high frequencies [10]. However, the performance of SRR-based metamaterials is limited at optical frequencies due to high conduction losses of the constitutive metallic elements, and the kinetic inductance of the electrons.

To improve the overall performance of metamaterials at optical frequencies, recently there was proposed an idea to employ dielectric particles with low or negligible losses in the visible spectrum. According to Mie theory, the scattering of electromagnetic waves by a small non-magnetic dielectric particle with high permittivity may

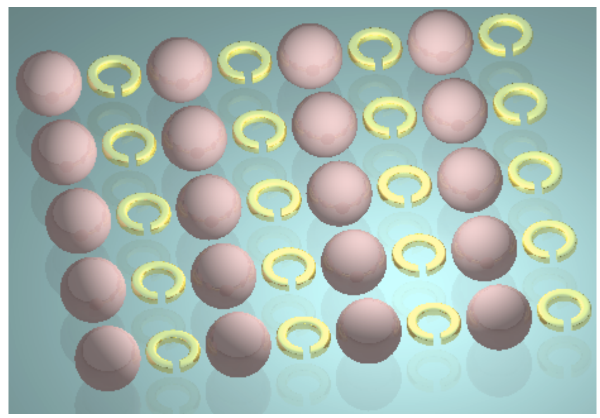

FIG. 1. (Color online) Schematic of a hybrid metal-dielectric structure consisting of silicon spheres coupled to copper split-ring resonators.

exhibit a strong magnetic resonance [11]. The induced magnetic response is connected to the excitation of localized modes of a dielectric particle for which the radial component of the magnetic field does not vanish. In this case, a dielectric particle can be considered as a magnetic-dipole scatterer of an incident electromagnetic wave at the resonant wavelength. This magnetic resonance results in an increased magnetic field in the near-field region around the particle. Recently, it was demonstrated that a silicon sphere with the radius in the range of $R = 40 - 100$ nm can have a magnetic resonance located in the visible spectrum [12]. It makes such particles the best candidates to achieve low-loss magnetic response at high frequencies.

In this Letter, we take one step further in the manipulation and control of the magnetic response and demonstrate that combining the magnetic resonances of silicon spheres and split-ring resonators (see Fig. 1) would allow to achieve a strong *antiferromagnetic response* by placing them in a close proximity. We expect that onedimensional periodic arrays and two-dimensional periodic lattices of such hybrid elements will support novel types of ferromagnetic and antiferromagnetic spin waves. This approach may open novel, yet unexpected ways for manipulating an optically-induced magnetic response of metamaterials in high-frequency range.

General concept and formalism. In the long-wavelength limit, when the characteristic sizes of a dielectric sphere and SRR are much smaller than the wavelength of the incident light, namely $r_{\mathrm{S,SRR}} \ll \lambda$, their optical properties can be described by the effective dipoles

$$\mathbf{p}^{\mathrm{S}} = \epsilon_0 \alpha^{\mathrm{S}} \mathbf{E}_0 \ , \ \ \mathbf{m}^{\mathrm{S,SRR}} = \chi^{\mathrm{S,SRR}} \mathbf{H}_0 \ , \tag{1}$$

where $\mathbf{p}^{\mathrm{S}}$ and $\mathbf{m}^{\mathrm{S}}$ are the induced electric and magnetic dipoles of the dielectric sphere, $\mathbf{m}^{\mathrm{SRR}}$ is the magnetic dipole of SRR, α_{S}, χ_{S}, χ_{SRR} are the electric and magnetic polarisabilities, $\mathbf{E}_0$ and $\mathbf{H}_0$ are the incident electric and magnetic fields. Since the electric response of SRR is very weak, it can be omitted.

The polarisabilities of the sphere can be obtained from the scattering dipole coefficients a_1 and b_1 of the exact Mie solution [11]

$$\alpha_E^{\mathrm{S}} = 6\pi i \frac{a_1}{k^3} \ , \chi_M^{\mathrm{S}} = 6\pi i \frac{b_1}{k^3} \ , \tag{2}$$

where $k = 2\pi/\lambda$ is a wavenumber, and the magnetic polarizability of SRR can be approximated as [9]

$$\chi^{\mathrm{SRR}} = \frac{F\omega^2}{(\omega_0^2 - \omega^2)} \ , \tag{3}$$

where ω_0 is the resonant frequency of a single SRR, and F is a geometrical factor.

To study the optical response of a pair of interacting dielectric sphere and SRR, we employ the method of coupled electric and magnetic dipoles [13–15] and we write the following system of equations

$$\begin{aligned}
\mathbf{p}^{\mathrm{S}} &= \alpha^{\mathrm{S}} \left\{ \epsilon_0 \mathbf{E}_0 - \frac{d}{c} (\mathbf{n} \times \mathbf{m}^{\mathrm{SRR}}) \right\} \ , \\
\mathbf{m}^{\mathrm{S}} &= \chi^{\mathrm{S}} \left\{ \mathbf{H}_0 + a\mathbf{m}^{\mathrm{SRR}} + b(\mathbf{n} \cdot \mathbf{m}^{\mathrm{SRR}})\mathbf{n} \right\} \ , \\
\mathbf{m}^{\mathrm{SRR}} &= \chi^{\mathrm{SRR}} \left\{ \mathbf{H}_0 + a\mathbf{m}^{\mathrm{S}} + b(\mathbf{n} \cdot \mathbf{m}^{\mathrm{S}})\mathbf{n} - dc(\mathbf{n} \times \mathbf{p}^{\mathrm{S}}) \right\} \ ,
\end{aligned} \tag{4}$$

where $c = 1/\sqrt{\epsilon_0 \mu_0}$ is the speed of light, $\mathbf{n}$ is a normal vector pointing from the sphere to SRR, and the coefficients a, b, and d describe the interaction of dipoles [16]

$$\begin{aligned}
a &= \frac{e^{ikR}}{4\pi R} \left(k^2 - \frac{1}{R^2} + \frac{ik}{R} \right) \ , \\
b &= \frac{e^{ikR}}{4\pi R} \left(-k^2 + \frac{3}{R^2} - \frac{3ik}{R} \right) \ , \\
d &= \frac{e^{ikR}}{4\pi R} \left(k^2 + \frac{ik}{R} \right) \ ,
\end{aligned} \tag{5}$$

$$\tag{6}$$

where R is the distance between the sphere and SRR.

For a sphere and SRR being aligned along the x-axis [$\mathbf{n} = (1, 0, 0)$], and the incident light propagating along the z direction [$\mathbf{k} = (0, 0, k)$], polarized with the magnetic field along the y-axis [$\mathbf{H}_0 = (0, H_y, 0)$], the solution of the system can be written as follows

$$\begin{aligned}
m_y^{\mathrm{S}} &= \frac{(1 + a\chi^{\mathrm{SRR}} + d^2 \alpha^{\mathrm{S}} \chi^{\mathrm{SRR}})}{(1 - a^2 \chi^{\mathrm{S}} \chi^{\mathrm{SRR}} + d^2 \alpha^{\mathrm{S}} \chi^{\mathrm{SRR}})} \chi^{\mathrm{S}} H_y \ , \\
m_y^{\mathrm{SRR}} &= \frac{(1 + a\chi^{\mathrm{S}})}{(1 - a^2 \chi^{\mathrm{S}} \chi^{\mathrm{SRR}} + d^2 \alpha^{\mathrm{S}} \chi^{\mathrm{SRR}})} \chi^{\mathrm{SRR}} H_y \ , \\
p_x^{\mathrm{S}} &= \epsilon_0 \alpha^{\mathrm{S}} E_x \ , \quad p_z^{\mathrm{S}} = -\left(\frac{d}{c} \right) \alpha^{\mathrm{S}} m_y^{\mathrm{SRR}} \ ,
\end{aligned}$$

with all other components vanishing. This simple solution gives us an useful insight and provides the information about the optical response of the hybrid system in different limiting cases. For the case of far separated sphere and SRR, their magnetization will be identical to the uncoupled system since $a, d \to 0$ with $R \to \infty$, resulting in $m_y^{\mathrm{S,SRR}} = \chi^{\mathrm{S,SRR}} H_y$. However, in the opposite limit when the sphere and SRR are placed very close to each other ($R \to 0$), the situation changes dramatically. The electric dipole moment of the sphere contributes strongly to the magnetization of the sphere itself since the cross-coupling coefficient $d^2 \propto R^{-4}$ becomes larger in the leading order compared to the self-coefficient $a \propto R^{-3}$ for very small distances $R \to 0$. This self-action of the longitudinal electric dipole on the sphere magnetization may result in the opposite magnetic response compared to the case of the uncoupled pair, depending of the relative signs of α^{S} and χ^{SSR}, keeping the direction of the magnetization of the SRR element unchanged. Thus, by placing a sphere and SRR in a close proximity, we may expect the appearance of the *antiferromagnetic response* of the whole structure.

Magnetization of a single meta-molecule. Based on the general analysis presented above, we consider a small silicon sphere of radius $R = 150$ nm interacting with a multilayer stack of identical copper SRRs with inner radius $R_1 = 70$ nm and outer radius $R_2 = 75$ nm, thickness $h = 5$ nm, and the gap width $g = 10$ nm. We use the multi-stack of SRRs with five layers to enhance their collective magnetic response. For the given parameters, the decoupled dielectric sphere and split-ring resonator have close magnetic resonances $\omega_{\mathrm{S}} = 281$ THz and $\omega_{\mathrm{SRR}} = 250$ THz, respectively [see Fig. 2(a)]. By placing the sphere and SRR in a close proximity, we may expect an effective magnetic coupling between the elements of different origin.

Our numerical simulations based on CST Microwave Studio confirm directly the existence of a strong optically-induced magnetic coupling in the hybrid metamolecule. We observe that, in addition to redshifting independent magnetic resonances of the sphere and SRRs where the magnetic response is dominated by one or another component, there appears a novel, *antiferromag-*

netic response with anti-parallel magnetization of both the elements of comparable strength in the vicinity of the SRRs magnetic resonance $\omega_{\rm AFM} = 257$ THz. Our analysis suggests that the longitudinal electric dipole moment of the sphere induced by SRRs plays an important role in the flip of the sign of magnetization, when two elements are placed in a close proximity. Indeed, by studying the dependence of the magnetization on the distance between two elements, as shown in Fig. 2(b), one can clearly see that both elements exhibit a ferromagnetic-like response for larger distances $R > R_{\rm cr}$, which changes to the antiferromagnetic-like response for smaller distances $R < R_{\rm cr}$, with opposite magnetization. This structure suggests that for optimal anti-ferromagnetic response the basic meta-molecule should consist of different constitutive elements to impose different mutual feedbacks.

Magnetic response of a metamaterial. Next, we consider a two-dimensional lattice of such hybrid meta-molecules which correspond to a hybrid metamaterial. The antiferromagnetic response of a single element implies that the periodic structure may also exhibit the staggered magnetization in the transverse direction. The latter is of a resonant origin, and it is closely related to the antiferromagnetic response. In Fig. 3(a) we plot the dispersion diagram of two-dimensional lattice of coupled system of hybrid meta-molecules with a period $D_z = 350$ nm in the z-direction, and compare it with the dispersions of the corresponding lattices of spheres and SRRs with the same period. We observe clearly the formation of a new branch in the coupled system corresponding to the staggered antiferromagnetic-like magnetization in the transverse direction. But, in the vicinity of the band edge we may also expect a staggered magnetic response in the longitudinal direction too, resulting in checker-board pattern of the total magnetization of the two-dimensional structure. There could be several reasons for that, including: (i) staggered induced magnetic field along the propagation at the band edge, and (ii) interaction of induced longitudinal electric dipoles of dielectric spheres. Both such conditions are met for the antiferromagnetic branch near the frequency $\omega = 260$ THz, which corresponds to the antiferromagnetic resonance of a single meta-molecule [see Fig. 2(a)] and the band edge of the two-dimensional structure [see Fig. 3(a)]. In Fig. 3(c) we show numerical results for the excitation at this particular frequency, which confirm the possibility of antiferromagnetic excitation in two directions with nearly ideal staggered magnetization. Moreover, this frequency corresponds to the crossing of two dispersion curves, where mostly dielectric spheres are excited. Thus, it leads to stronger magnetic interaction between spheres and SRRs, where the resonantly excited longitudinal dielectric dipoles of the spheres will play an essential role for magnetization along the propagation. We find that robust staggered magnetization pattern of dielectric spheres weakly depends on the periodicity in the $z-$ direction up to the values of $D_z = 600$nm for frequencies in the vicinity of the antiferromagnetic resonance of a single hybrid element [see Fig. 2(a)]. For lager periods, the dispersion diagram of the coupled systems changes significantly, and the antiferromagnetic branch shifts away from that resonant frequency.

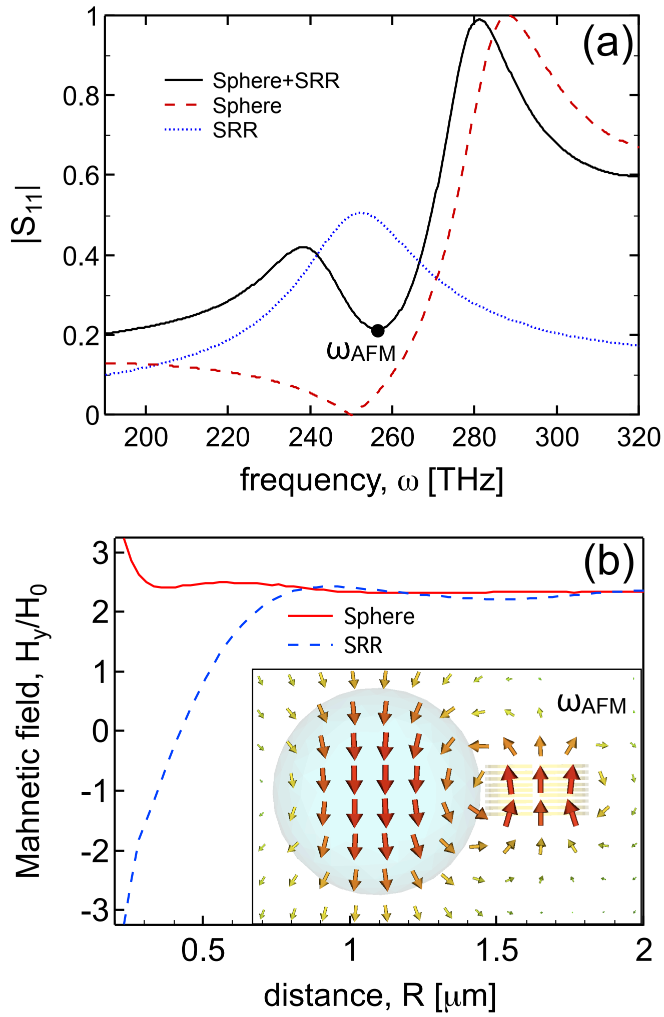

FIG. 2. Magnetic response of a silicon sphere coupled to a multi-stack of SRRs. (a) Reflection curves of single elements [single silicon sphere (dashed line) and multi-stack of SRRs (dotted line)] , and of the coupled structure (solid line). Two peaks of the coupled system correspond to the magnetic resonances of isolated elements. Resonant dip corresponds to the anti-ferromagnetic like excitation with staggered magnetic polarisabilities; (b) dependence of the induced magnetization of the sphere (solid line) and SRRs (dashed line) on the distance between at the resonant frequency $\omega_{\rm AMF} = 257$ THz. Below critical distance $R_{\rm cr} \approx 1\mu m$ the ferromagnetic like magnetization switches to anti-ferromagnetic one; Insert shows the distribution of the induced magnetic field at the antiferromagnetic resonance.

To estimate the propagation length of the induced spin waves in such a structure, we employ the method recently suggested in Ref. [17], which allows us to extract complex

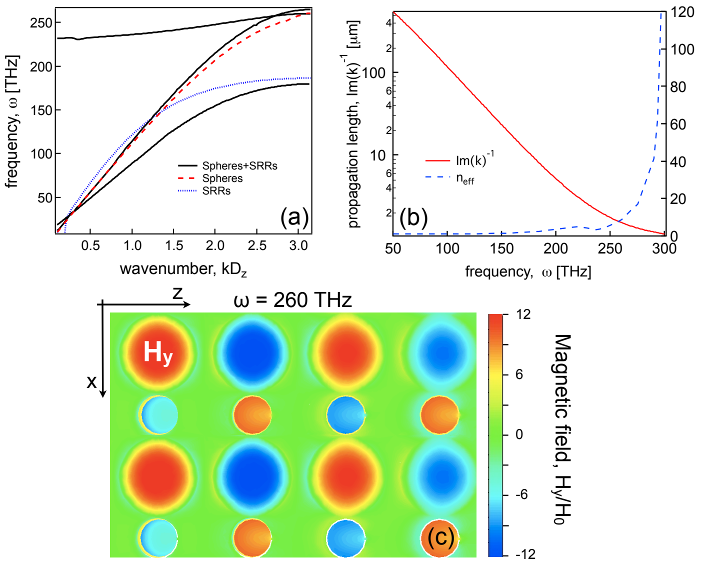

FIG. 3. (Color online) (a) Comparison of the dispersion diagrams of two-dimensional lattices of spheres (dashed line), SRRs (dotted line), and hybrid sphere-SRR system (solid lines) with the periodicity in z-direction $D_z = 350$ nm; (b) Calculated propagation length (solid line) and effective refractive index (dashed line) of a finite structure; (c) Magnetic field distribution H_y of two-dimensional structure at the resonant frequency $\omega = 260$ THz. This gives a typical example of the staggered two-dimensional checker-board magnetization of the hybrid structure.

propagation constants of the Bloch modes based on the finite number sample points per period. The results are summarized in Fig. 3(c), where the propagation length is inversely proportional to the imaginary part of the corresponding wavevector, $\sim \mathrm{Im}(k)^{-1}$. It decays exponentially with frequency, and the antiferromagnetic resonance $\omega_{\mathrm{AFM}} = 257$ THz corresponds to six periods of the structure. At the same time, the effective refractive index becomes relatively large, $n_{\mathrm{eff}} \approx 10$, with slow phase velocity. One of the reasons is that this particular resonant frequency corresponds to the band edge of the antiferromagnetic mode [see Fig. 3(a)], which can be altered with the proper period in the z-direction.

In conclusion, we have studied the optically-induced magnetic response of hybrid metal-dielectric structures consisting of dielectric spheres and multilayer stacks of split-ring resonators. We have revealed that each element of such a structure exhibits a strong magnetic response in the THz frequency range, and coupling of two different elements leads to a strong magnetic interaction between the spheres and split-ring resonators. Based on the coupled-dipole model we have demonstrated an important role of resonantly-induced longitudinal electric dipoles in the formation of an antiferromagnetic response with a staggered magnetization pattern. Arranging the hybrid meta-atoms into a two-dimensional lattice allows to create a hybrid metamaterial supporting staggered checker-board magnetization patterns in two directions due to the magnetic interaction between dielectric spheres. A weak dependence of the staggered magnetization of the dielectric spheres on the distance between them at the antiferromagnetic resonance of a single meta-molecule indicates a significant contribution of the induced longitudinal electric dipoles into the overall magnetic interaction. We believe this approach opens novel possibilities for the manipulation and control of artificial antiferromagnetism at optical frequencies.

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