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PAPER

# Antiferromagnetic order in hybrid electromagnetic metamaterials

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## Abstract

We demonstrate experimentally a new type of order in optical magnetism resembling the staggered structure of spins in antiferromagnetic ordered materials. We study hybrid electromagnetic metasurfaces created by assembling hybrid meta-atoms formed by metallic split-ring resonators and dielectric particles with a high refractive index, both supporting optically-induced magnetic dipole resonances of different origin. Each pair (or 'metamolecule') is characterized by two interacting magnetic dipole moments with the distance-dependent magnetization resembling the spin exchange interaction in magnetic materials. By directly mapping the structure of the electromagnetic fields, we demonstrate experimentally that strong coupling between the optically-induced magnetic moments of different origin can flip the magnetisation orientation in a metamolecule creating an antiferromagnetic lattice of staggered optically-induced magnetic moments in hybrid metasurfaces.

Natural materials are known to exhibit negligible magnetism at optical frequencies, since the direct effects of the optical magnetic field on matter are much weaker than electric ones. These restrictions can be overtaken by the concept of metamaterials created as artificial structures of subwavelength meta-atoms that support a strong optically induced magnetic response [1]. The recent progress in the physics of metamaterials and their applications for metadevices [2] is based on their ability to achieve unprecedented control of optical properties in artificial electromagnetic structures.

After the first experimental verification of exotic properties of metamaterials such as superlens with the resolution beyond the diffractive limit [3] and electromagnetic cloaking [4], the metamaterials paved a way for the realization of a new generation of microwave and optical metadevices [2] with numerous unique functionalities and many potential applications [5, 6]. Moreover, the concept of metamaterials and metasurfaces now spans over various branches of physics, including acoustics, mechanics, superconductors, and advanced bio-engineering. Based on conventional materials, metamaterials derive their unique properties from their newly designed structures.

It is known that natural materials can exhibit various dielectric permittivity at optical frequencies. In contrast, the magnetic permeability is always close to its free space value in the optical range. Thus, in all conventional optical materials and devices, only the electric component of light is directly controlled. However, the emergence of metamaterials has fundamentally altered this situation. Artificial magnetism can now be achieved at higher frequencies by engineering specially designed 'meta-atoms'—functional units of metamaterials that are smaller than the wavelength and carry an optically induced magnetic moment.

When such meta-atoms are combined to create a metamaterial or metasurface, the overall structure of the induced magnetic moments resemble the ferromagnetic (FM) order, as shown in two cases in the upper part of figure 1. This is valid for both types of magnetic moments, either induced in metallic meta-atoms by the oscillation of free electrons or by displacement current in dielectric meta-atoms with high refractive index. However, if we assemble hybrid structures with dissimilar elements having the magnetic dipole moments of

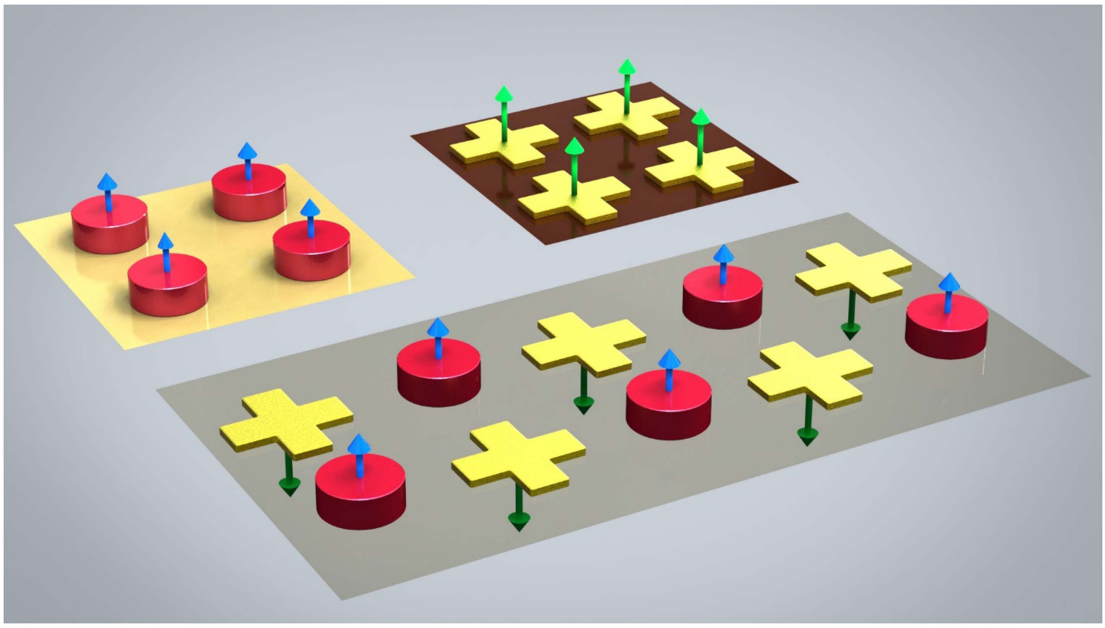

**Figure 1.** Schematic of the magnetic dipole ordering in electromagnetic metasurfaces. On the upper part two generic metasurfaces composed of the arrays of different elements (meta-atoms) exhibit a FM or AFM ordering depending on the distance between elements, when magnetic dipole moments are induced. Being combined together, two dissimilar types of meta-atoms with the FM ordering can create an AFM ordering with the staggered structure of optically-induced magnetic dipole moments. The structure is one of several possible realizations of the AFM order. The other type of element ordering is shown e.g. in figure 1 in [7].

different origin (see the lower part of figure 1), we expect that such hybrid metamolecule with a strong coupling of individual moments can create an antiferromagnetic (AFM) order of magnetic moments, as was recently suggested theoretically [7]. The experimental verification of this concept and the demonstration of the AFM ordering for a single metamolecule and even full metasurfaces is the main subject of this paper.

One of the canonical subwavelength 'meta-atom' is a split ring resonator (SRR) that consists of an inductive metallic ring with a gap. The basic principle behind this design is that SRR can support the principal eigenmodes with a circular current distribution that gives rise to an induced magnetic moment and, after assembling such elements into a structure with the period ordering, to the optical magnetism, the cornerstone concept of the modern theory of metamaterials. Metamaterials composed of regular lattices of SRRs have been studied in a significant number of papers. If we look at all those structures from the viewpoint of magnetism and magnetic systems, we should identify the order of magnetic moment as ferromagnetic, when all moments are predominantly pointed to a certain direction only.

Importantly, the intrinsic losses put a fundamental limit for downscaling the SRRs for obtaining an efficient magnetic response in the optical range. On the other hand, resonant dielectric nanostructures make a new twist on light–matter interaction [8]. According to the analysis employing Mie's scattering theory [9, 10] and recent experimental verifications [10, 11], subwavelength dielectric nanoparticles with high refractive index may exhibit very strong magnetic response in the entire visible spectral range. The basic physical mechanism of the induced optical magnetism in such a dielectric particle is the excitation of a particular mode inside the particle with a circular displacement current of the electric field. The spectral position of the magnetic resonance can be tuned throughout the whole visible spectral range from violet to red by changing the nanoparticle size in the range from 100 to 200 nm [9, 11]. As a result, it makes silicon nanoparticles the best candidates for lossless magnetic response at high frequencies. One of their most successful applications is the superior performance of hybrid and all-dielectric nanoantennas for effective control of light emission from localized sources [13–19]. Strong near-field enhancement in resonant dielectric nanostructures [20, 21] implies that strong nonlinear phenomena can be expected in the nonlinear optical response of dielectric nanoparticles with high refractive index. Recently, the enhancement of the third-order optical nonlinearities of silicon nanodisks has been observed at the vicinity of the magnetic dipole resonances pumped by femtosecond laser pulses [22]. The efficiency of the conversion is enhanced by two orders of magnitude with respect to the unstructured bulk silicon slab. These results demonstrate a possibility of establishing a novel efficient platform of nanoscale resonant nonlinear optics driven by an optically-induced magnetic response of low-loss high-index nanoparticles. Moreover, the regular arrays of such bi-spherical elements would allow the development of all-dielectric optical 3D metamaterials.

Combining both metallic and dielectric meta-atoms allows to create hybrid metamaterials taking the advantages of both plasmonic and all-dielectric elements. In particular, such structures may offer large Purcell enhancements and highly directional radiation patterns while preserving high radiation efficiencies [16, 17]. However, the current study of combined effects of metallic and dielectric hybrid nanostructures on the magnetic nanostructures is very limited, and it is mainly restricted by the radiation control [12].

In this paper, we provide the first experimental evidence of a new type of optically-induced magnetic ordering in hybrid metamaterials resembling staggered structures of spin in antiferromagnetic materials. We experimentally study metamaterials created by assembling periodically hybrid metamolecules supporting the interaction of magnetic dipole moments of different origin. We demonstrate that in the regime of strong coupling between the magnetic moments one can observe the change of the magnetisation orientation to AFM type ordering depending on the separation distance between the particles.

## Results and discussion

Based on the theoretical suggestion [7], we consider the optically-induced interaction of magnetic moments in hybrid structures consisting of a dielectric sphere and a split-ring resonator shown to drastically change the magnetic response due to the different origin of their magnetic dipole resonances: one is based on the inductive response of normal metals in SRRs, and the other one based on the displacement current inside the dielectric particle. In particular, it was predicted [7] that the resonantly induced longitudinal electric dipoles play an important role in the formation of a staggered magnetization with the AFM response below a critical separation between the metallic and dielectric elements of the hybrid metamolecule.

First, we optimise the theoretical results following closely the analysis presented in [7] for a single hybrid metamolecule made of a dielectric sphere with permittivity $\varepsilon = 16 + 0.001\mathrm{i}$ and the radius $R_1 = 7.5$ mm and a copper SRR ring with the radius $R_2 = 5.3$ mm and thickness 0.05 mm (see figure 2). These material and geometrical parameters are linked closely to the available samples and materials for our experimental verification summarized below. Figure 2 clearly demonstrates and confirms that below a critical separation between two simple meta-atoms, namely around $d_c = 30$ mm, the initial in-phase excitation of magnetic dipole moments flips to the staggered configuration, in the vicinity of the magnetic dipole resonance at 4.73 GHz. We notice that the incident plane wave propagates perpendicular to the axis of the hybrid metamolecule with the electric field polarized along the axis. Since the excitation is kept the same, the flipping of the magnetic dipole moment is attributed to the optically-induced interaction of two meta-atoms. Interestingly at critical distance $d_c = 30$ mm both induced magnetic dipoles are orthogonal to each other, thus at any given moment of time (or phase) only one of them is effectively aligned along the applied magnetic field, while another one is across the field. The situation changes over quarter of a period. It is crucially important that this type of interaction may only occur for magnetic dipoles.

Next, we move to the experimental verification of the existence of the AFM ordering. We experimentally study the properties of a single hybrid metamolecule made of ceramic MgO-$TiO_2$ sphere with the parameters closed to those simulated numerically and described above. In figure 3 we compare the theoretical and experimental data for the scattering cross sections for a single SRR and ceramic sphere, as well as for a hybrid metamolecule, for the separation below critical value $d_c$ (where $d_c = 30$ mm), and observe that they are in a very good agreement. Both magnetic resonances of individual meta-atoms are spectrally close to each other being located at 4.59 GHz and 4.8 GHz for SRR and dielectric sphere, respectively. Importantly, the AFM ordering is associated with resonant suppression of the total scattering cross section. This can be understood due to the out-of-phase excited magnetic dipoles resulting in scattering cancelation in the far-field. Such a behaviour can be interpreted also in terms of the Fano resonances [23, 24], where the SRR makes a resonant contribution with the $\pi$-phase shift of the magnetic dipole, and the dielectric particle provides the scattering background. Since the symmetry of the radiation pattern of both modes are identical, they lead to destructive interference in the far-field zone, similar to recently demonstrated nonradiating anapole modes [25].

In figures 4(a) and (c) we show the phase distribution of the magnetic field just above the hybrid metamolecule along its axis. Both numerical and experimental results exhibit sharp phase variation in the vicinity and in-between of individual magnetic dipole resonances. In figures 4(b) and (d) we plot the phase distribution at the antiferromagnetic resonance in the plane on top of the hybrid metamolecule. It clearly shows out-of-phase magnetic field distribution in the vicinity of SRR and dielectric particle.

Finally, we consider the periodic arrangement of hybrid metamolecules in a two-dimensional square lattice. In general, there are several different possible ordered arrangements. For example, the magnetic moments of the hybrid metamolecules alternate only in one direction remaining identical in the other direction. Another possibility is the arrangements of magnetic moment of hybrid metamolecules alternate in both directions, similar to the check-board pattern (see figure 5). Such hybrid composite metasurfaces may support optically-

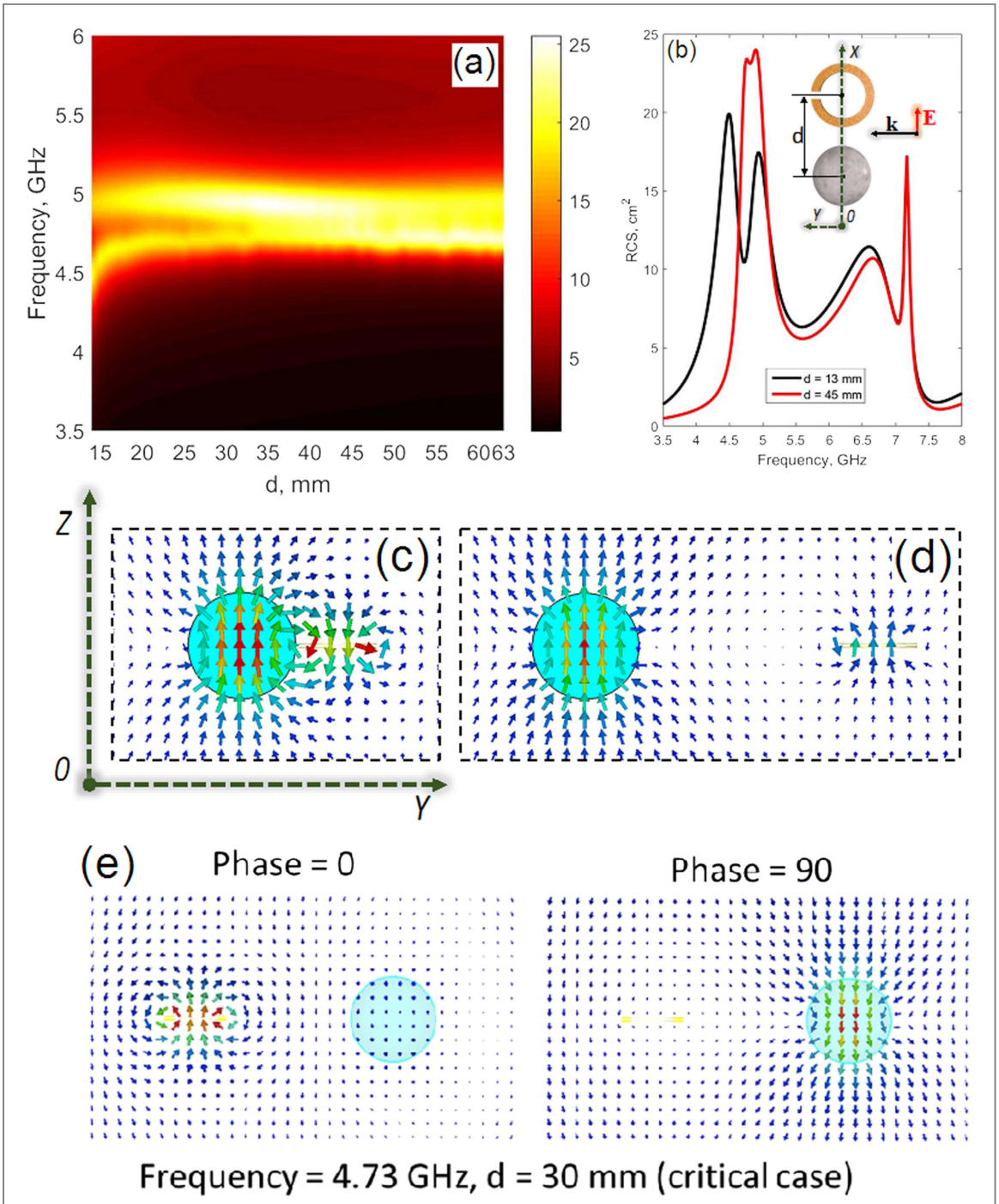


**Figure 2.** Scattering cross section of a hybrid metamolecule. (a) Two-dimensional plot of the scattering cross section characterising the magnetic interaction in the hybrid metamolecule versus the separation between two constituent meta-atoms. (b) Characteristic scattering cross sections above ($d = 45$ mm) and below ($d = 13$ mm) the critical coupling, exhibiting the resonant suppression of the total scattering for the AFM ordering. (c), (d) Magnetic field distribution showing a transition from FM to AFM type ordering at the resonant frequency 4.73 GHz for the separation $d = 13$ mm and $d = 45$ mm, respectively. (e) Magnetic field distribution at critical coupling $d = 30$ mm showing that resonant magnetic dipoles are present within both elements, but they rotated 90 degrees with respect to each other, which leads to phase shifted excitation.

induced magnetic response characterized by the staggered magnetization in the longitudinal and transverse directions due to the magnetic interaction between the dielectric particles. In figure 5(b) we show our experimental results for a finite structure consisting of closely packed ceramic spheres and copper SRRs. A direct phase measurement reveals the staggered magnetic field distribution above the metasurface, in a full agreement with our numerical simulation, shown in the top panel of figure 5(a).

For comparison, we also perform the numerical simulations for metasurfaces made of identical meta-atoms (either SRRs or dielectric particles). It turns out that, at the magnetic dipole resonance, such structures exhibit only ordered states that resemble the familiar FM ordering of spins in magnetic materials. This finding indicates the importance of the different origin of the magnetic dipole response. In particular, as was demonstrated

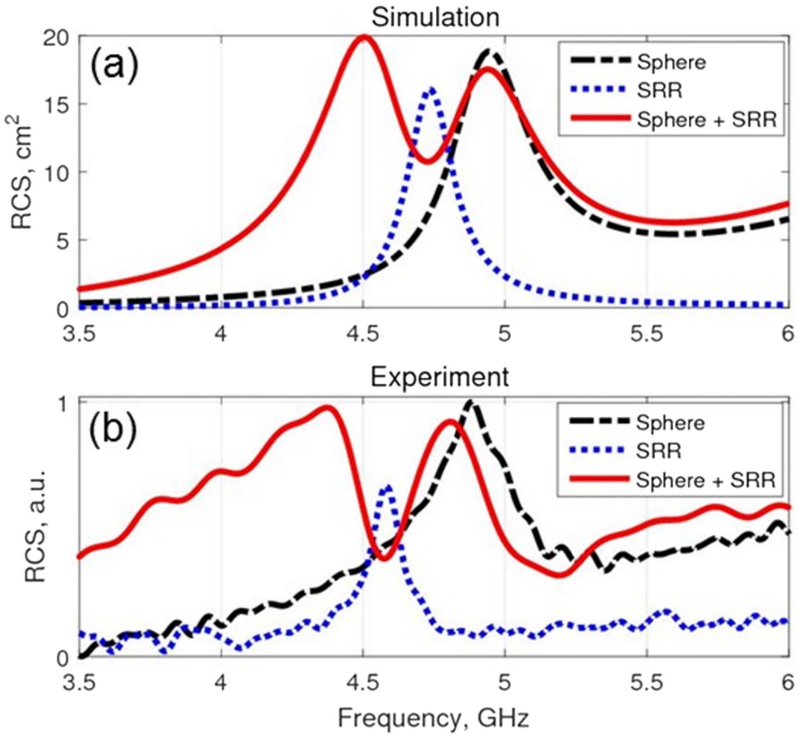


**Figure 3.** Scattering cross sections of individual meta-atoms and hybrid metamolecule. Individual meta-atoms (SRR and sphere) exhibit strong magnetic dipole resonances shown by dotted (SRR) and dashed (sphere) curves, as observed in (a) numerical simulations and (b) experiment. Being combined into the hybrid metamolecule, the magnetic response demonstrates a splitting of resonances responsible for the formation of the AFM order.

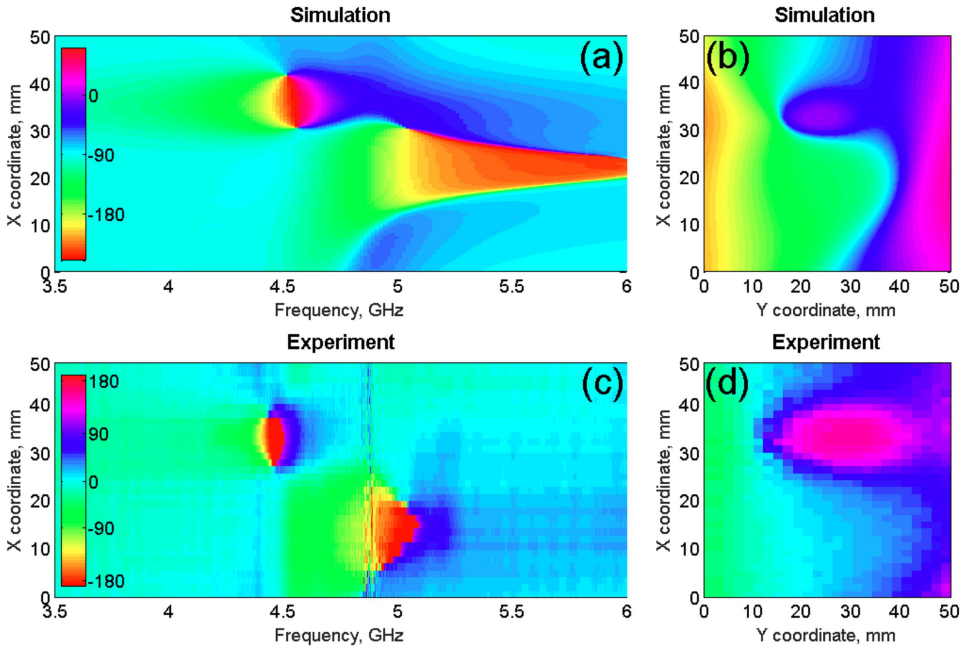


**Figure 4.** Phase structure of a single hybrid metamolecule. (a), (c) Spectral variation of the magnetic field phase above the hybrid metamolecule along the OX line indicated in figure 2(b), calculated theoretically and measured experimentally. (b), (d) Two-dimensional phase distribution in the plane on top of the hybrid metamolecule at the distance 1 mm above and the resonant frequency 4.73 GHz.

analytically in [20], one of the important features responsible for the formation of the AFM order is the existence of both electric and magnetic dipole resonances supported by the dielectric particle. As was recently demonstrated, both types of response can be controlled independently by altering the shape of the dielectric

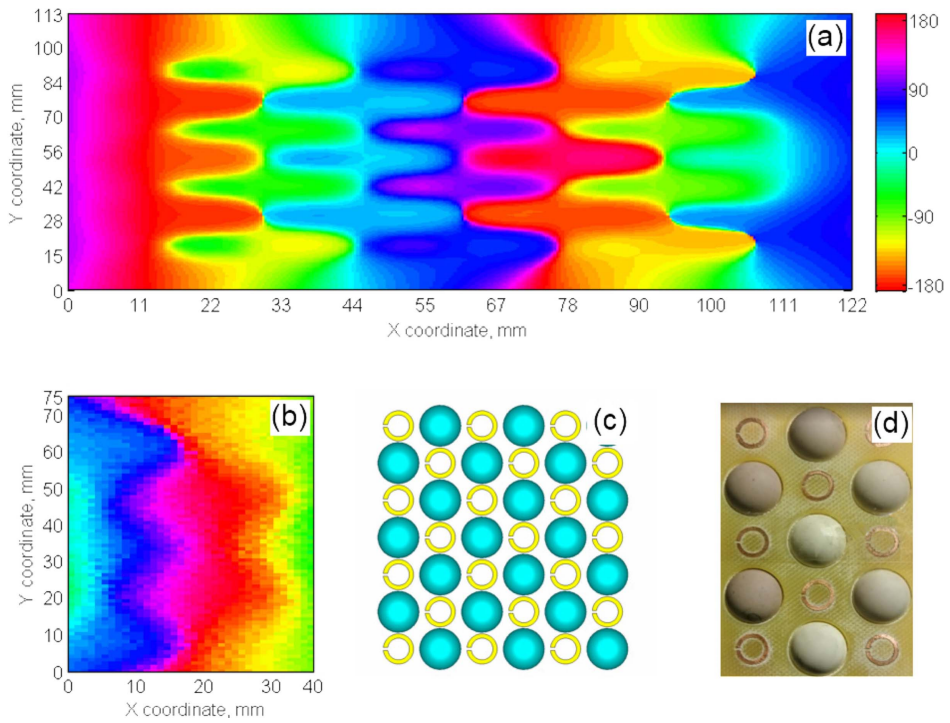


**Figure 5.** Magnetic field phase distribution of a hybrid metamaterial. (a), (b) Theoretical and experimental phase distribution of the magnetic field 1 mm above the hybrid metamaterial at the frequency 4.73 GHz. (c) Schematic view of the simulated structure and (d) camera photo of the finite experimental sample.

particle [26]. It opens new opportunities to fine-tuning and controlling the AFM type ordering of optically induced magnetic dipole moments.

AFM metamaterials closely resemble real AFM materials in nature which are internally magnetic, but have resulting zero net magnetic moment. This implies that information stored in antiferromagnetic moments would be invisible to common magnetic probes, insensitive to disturbing magnetic fields, and the antiferromagnetic element would not magnetically affect its neighbours, regardless of how densely the elements are arranged in the device. Alternatively, it is possible to consider a triangular lattice of such hybrid metamolecules which will resemble the graphene-type honeycomb lattice. It is expected that AFM order might lead to formation of topologically nontrivial helical edge states similar to quantum spin-Hall effect. Moreover, we expect many interesting problems to be solved here, in analogy with on recent progress of the field of AFM spintronics for spin transport [27], magnetic textures, spin dynamics, and free electron lasers.

In summary, we have demonstrated experimentally the novel antiferromagnetic type ordering of magnetic moments in hybrid metamaterials consisting of both metallic and dielectric components. Our analysis suggests that such effect is based on the properties of a single hybrid metamolecule due to a combination of elements with different origins of optically-induced magnetic dipole response. By changing the geometry of the dielectric particle it is possible to tune independently electric and magnetic type responses [26]. Such an approach can be further extended to achieve fine control of magnetic interaction with spectral tuning of multipolar coupling of different modes. Finally, hybrid metamaterials offer new opportunities to achieve unprecedented control of light–matter interaction.

## Methods

### Experimental approach

For our microwave experiments, we employ MgO-$TiO_2$ ceramic spheres characterized by a dielectric constant of 16 and dielectric loss factor of $(1.12–1.17) \times 10^{-4}$, measured at 3–9 GHz frequency range. To fasten together the particles, we use a special holder made of a Styrofoam material with dielectric permittivity of 1 (in the microwave frequency range). To realize a plane wave excitation, we employ a rectangular horn antenna (TRIM 0.75–9 GHz; DR) connected to the transmitting port of a vector network analyzer (Agilent E8362C). For the measurements of the scattering cross section, an isolated hybrid metamolecule is placed in the far-field zone of the antenna at the distance approximately ~2.5 m, and the second horn antenna (TRIM 0.75–9 GHz; DR) is used as a receiver. In accord with the optical theorem, the scattering in the forward direction is connected with the total extinction

cross section. Since absorption by dielectric material is negligible, the total extinction cross section coincides with the scattering cross section.

The near-field measurement scanning is performed for the frequencies within the 3–7 GHz frequency band. We use an automatic mechanical near-field scanning device and a shielded loop probe connected to the receiving port of the Analyzer. The shielded loop had a ~6 mm diameter, and it could be approximated as an electrically small current loop that measures the magnetic field in the forward direction without significant field averaging. The probe was oriented normally with respect to the interfaces of both meta-atoms and hybrid metamaterial as well as to the transmission horn antenna (dominantly measured H-component of the horn). The near-field mapping is scanned at the distance of ~1 mm away from the back interface of the metamaterial to avoid touching of the probe and sample.

### Numerical simulations

All numerical simulations for the scattering cross sections and near-field distributions are performed by using the commercially available CST Microwave Studio software. The permittivity is taken from a standard library. The permittivity of ceramic MgO-$TiO_2$ is retrieved from the experimental data and approximated by a constant value $\varepsilon = 16 + 0.001i$, for the given frequency range.

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