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Metamaterials with Tailorable Nonlinear Optical Properties

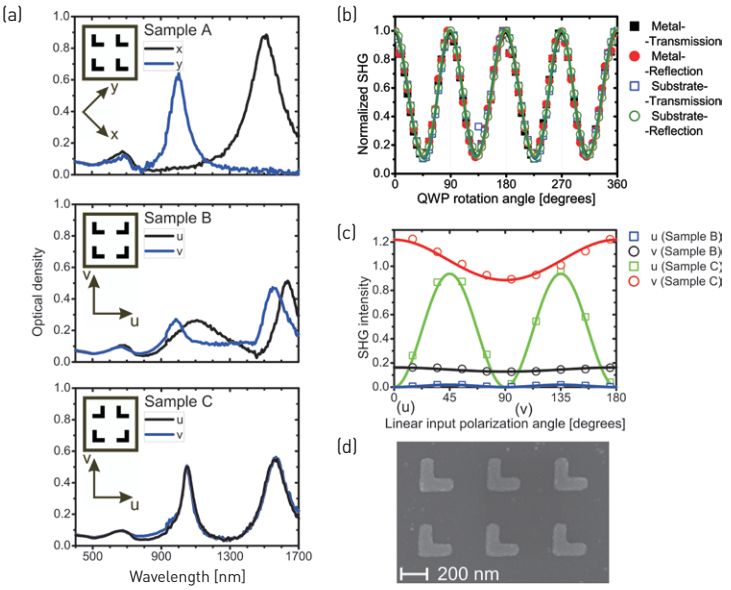
Second-order nonlinear processes such as second-harmonic generation (SHG) require noncentrosymmetric structures. The development of second-order metamaterials, however, has been hampered by symmetry breaking due to sample defects and shape distortions. The resulting outcomes can be interpreted in terms of effective higher-multipole (magnetic and quadrupole) effects that strongly modify the radiative properties of the samples.[1,2]

We have shown that samples with significantly improved quality suppress the higher multipole effects in SHG from arrays of metal nanoparticles. This is a prerequisite for developing nonlinear metamaterials with designed properties. To demonstrate this possibility, we have modified the SHG response by subtle details of particle ordering.

Our samples consist of arrays of L-shaped noncentrosymmetric gold nanoparticles fabricated by electron beam lithography and liftoff. They are arranged in a square array with a 500-nm period on top of a fused silica substrate. The high particle quality is already evident in their extinction spectra, which show narrow linewidths, indicating much less inhomogeneous broadening than earlier samples.

The role of defects and higher-multipole effects is addressed by comparing polarization-dependent reflected and transmitted SHG signals for the substrate and metal side incidence of the fundamental beam. In essence, interference between multipoles leads to differences in the four signals.[2,3] All signals for our samples overlap, showing that we have reached the dipole limit in the effective SHG response.[4] Additionally, SHG efficiency was enhanced by a factor of 10 compared to earlier samples.[3]

To tailor the SHG response, we fabricated samples where the relative orientations of the particles are varied in a 2 × 2 particle cell. All particles in reference sample A have the same orientation. Particles in samples B and C have two different orientations but these samples differ in detailed particle ordering. Surprisingly, the SHG responses of B and C differ by up to a factor of 50 for equivalent polarization-dependent signals, although both samples have identical orientational distribution of the particles.[5] Sample B is always inferior to the reference, whereas C is superior. These results can be understood in terms of long-range diffractive coupling between the particles, which becomes possible because the samples' unit cell size also depends on particle ordering. Our results have thus opened the way towards developing metamaterials with tailorable nonlinear properties. OPN



(a) Extinction spectra of different L-shaped nanoparticles arrays. Insets show the particle ordering in a 2 × 2 particle cell. (b) Normalized transmitted and reflected SHG signals from the reference sample A for metal and substrate incidence. (c) Normalized SHG intensity for samples B and C and the detected polarization components of the SHG intensities as a function of linear input polarization. (d) The SEM micrograph of the sample A.

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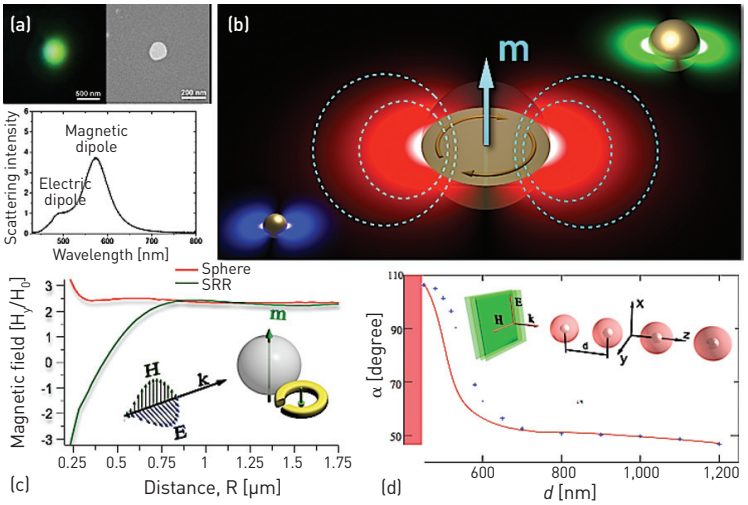
Magnetic Light: Optical Magnetism of Dielectric Nanoparticles

A split-ring resonator (SRR) is an inductive metallic ring with a gap that can support an oscillating current, giving rise to an optically induced magnetic moment. Unfortunately, metal intrinsic loss sets the limit for using SRRs at optical frequencies. But according to our work, spherical silicon nanoparticles can make an attractive alternative.

Dielectric nanoparticles exhibit strong magnetic resonances in the visible. Their excitation is similar to SRRs, but silicon nanoparticles have lower losses. Magnetic resonance originates from exciting an electromagnetic mode inside the nanoparticle with a circular displacement current of the electric field. This mode is excited when the wavelength is comparable to the particle's diameter. It has an antiparallel polarization at opposite ends of the particle while the magnetic field is oscillating up and down in the middle. We saw this fundamental phenomenon of strong magnetic resonances throughout the whole visible spectral range for silicon nanoparticles from 100 to 270 nm. Similar results have been observed in red and infrared spectral region.[1,2]

These dielectric nanoparticles could be used to explore nanoscale interactions. Coupling silicon nanoparticles and SRRs allows researchers to control the magnetic interaction between optically induced dipole moments. If the spacing between a nanoparticle and SRR becomes smaller than a critical value, the induced magnetization can be inverted. This leads to a staggered pattern of magnetic moments, with the potential for light-induced artificial antiferromagnetism at optical frequencies.[3] This approach can be generalized to create hybrid structures supporting and controlling optically induced spin waves.



(a) Close-view dark-field microscope (left) and SEM (right) images of a silicon nanoparticle with dark-field scattering spectra (bottom) exhibiting magnetic dipole (md) resonance. (b) Electric (yellow) and magnetic (blue) field distributions inside a high-refractive index dielectric nanoparticle at the magnetic resonance. (c) Dependence of the dielectric sphere and SRR optically induced magnetization on the separation distance at the magnetic resonance frequency. Below the critical distance, the optically induced ferromagnetic-like magnetization is replaced by an antiferromagnetic pattern. (d) Beam width of the radiation of core-shell nanoparticles vs. separation distance *d*.

The interaction between magnetic and electric dipoles may lead to new scattering properties. An interference between two optically induced dipole resonances results in azimuthally symmetric unidirectional scattering that can be realized in layered nanoparticles with metal cores and dielectric shells.[4] A superposition of electric and magnetic resonances of a single core-shell nanoparticle may give way to the suppression of backward scattering and unidirectional emission by a single subwavelength element.[4] Directivity can be enhanced by forming a chain of such nanoparticles. This work suggests a novel principle of optical nanoantennas made of dielectric nanoparticles.[5] These structures exhibit higher radiation efficiency than their plasmonic counterparts, allowing more compact designs. OPN

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