

# A Colloidal Solution of Crystalline Silicon Nanospheres: Toward Mie Resonance Based All Dielectric Metafluids

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## 1. Introduction

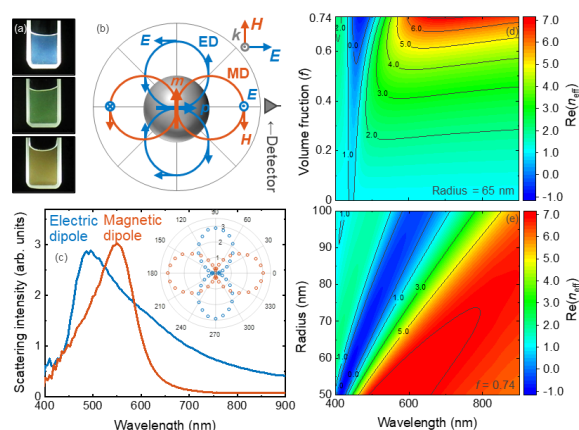
Meta-atoms that support magnetic and electric dipoles at visible frequencies are key building-blocks for optical metamaterials due to the emergence of the degree of freedom to tailor effective permittivity, permeability, and resultant refractive index. Recently, liquid-type metamaterials, solutions in which meta-atoms are dispersed, have been proposed as metafluids.[1,2] In contrast to solid-state metamaterials, the liquid metamaterials can be applied to solution-phase spectroscopy and optofluidic devices to enhance signals and/or offer advanced functionalities. As a component of liquid metamaterials, a plasmonic meta-atom composed of silica core decorated with metal nanoparticles have been studied so far; however, their magnetic responses are usually weaker than electric ones, limiting their tunability of macroscopic effective parameters. Also, the operation frequency is fixed at the localized surface plasmon resonance, and tuning the frequency is not straight forward.

Here, we propose an all dielectric metafluid consisting of silicon nanospheres as meta-atoms. The silicon nanosphere exhibit strong optical magnetism originating from Mie resonances without the requirement of complex structures unlike plasmonic counterparts. Using a recently developed colloidal solution of silicon nanospheres[3], we carry out macroscopic scattering measurement and observe strong magnetic responses with comparable strength to electric ones in bulk solutions for the first time. The strong and isotropic electric and magnetic responses allow us to control unnaturally high or low effective refractive indices over whole visible spectrum.

## 2. Results and discussion

The silicon nanospheres were colloiddally synthesized by a procedure recently reported.[3] Figure 1a is the picture of the synthesized colloidal solutions containing silicon nanospheres with different sizes. They exhibit blue, green, and yellowish colors due to the electric and magnetic dipole Mie resonances.

The contribution of the electric and magnetic dipoles can be separately measured using polarization-resolved scattering measurement using their orthogonal radiation patterns and polarizations of the scattered field as shown in Figure 1b. We illuminated white light from a halogen lamp through a polarizer and detected the scattered light at 90° against illumination direction. By rotating the polarizer and detecting at 90° and 0°, the electric and magnetic dipole scattering, respectively, can be separately measured. Figure 1c shows the scattering spectra of electric and magnetic



**Figure 1.** (a) Images of colloidal solutions of silicon nanospheres with 54, 65, and 99 nm in radii. (b) Radiation patterns of electric (blue) and magnetic (orange) dipole scattering. (c) Measured scattering spectra of electric and magnetic dipoles resonances. The inset shows the angular pattern of them. (d,e) Effective media calculation for a material consisting of silicon nanospheres for different (d) volume fraction and (e) radius.

dipole resonances measured for the green solution (radius = 65 nm). The peak intensities are comparable each other, suggesting the ideality of the solution for metafluids.

To show the possibility of silicon nanospheres as a metafluid, we calculate the effective refractive indices of dispersion of silicon nanospheres using the Maxwell-Garnett effective medium approximation into which the Mie theory is included (Figure 1d,e). As can be seen, the effective refractive index is dependent on the volume fraction, and near zero (<1) index can be obtained even in the low volume fraction. As a strong contrast to the plasmonic counterparts, the size dependence of the effective refractive index (Figure 1e) show the operation range (e.g., blue region) of the solution can be controlled over the entire visible spectrum. These results indicate the prominent possibility of proposed all-dielectric metafluid

## 3. Conclusions

The colloidal solution of silicon nanospheres are excellent candidate for a novel metafluid due to the strong electric and magnetic responses originating from Mie resonances. The metafluid may pave the way for optofluidic metamaterial devices.

## References

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- [3] H. Sugimoto et al., Adv. Opt. Mat. 202000033 (2020)