

# Two-photon reduction technique for isotropic metamaterials

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**Abstract**—The electromagnetic properties and design strategy of plasmonic metamaterials in the optical spectral range are theoretically investigated. The fabrication techniques for the three-dimensional isotropic metamaterials are also proposed. Three-dimensional nano-scale silver or gold structures are demonstrated.

**Keywords**—component; metamaterials; resonator; plasmonics; two-photon absorption; reduction; 3D structure

## I. INTRODUCTION

Plasmonic metamaterial is a structure that consists of metal resonator array. Designing resonator array structure so that it is smaller than the wavelength of the light, metamaterials works as a homogeneous material whose electromagnetic properties inherited from its structure. By engineering such materials, we can design and control their magnetic permeability even in the optical frequency region in which all materials in nature lose magnetic response and their relative permeability is fixed at unity. In this paper, we report on the theoretical investigations of the artificial magnetism of metamaterials in the visible light. Moreover, we also present a two-photon-induced metal-ion reduction technique in a metal-ion doped materials for fabricating 3D isotropic metamaterials.

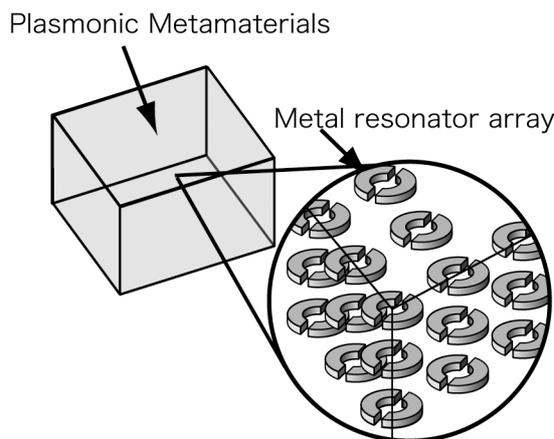


Figure 1 Plasmonic metamaterials.

## II. PLASMONIC METAMATERIALS AND THEIR EFFECTIVE PERMEABILITY

We have theoretically investigated the magnetic response of plasmonic metamaterials in the optical frequency region [1-4]. Figure 2 shows the split ring-resonator (SRR) model used in our calculations. Describing the dispersion properties of the metals in the optical region, we derived the effective permeability ( $\mu_{\text{eff}}$ ) of the SRRs as

$$\mu_{\text{eff}} = \mu_{\text{re}} + i\mu_{\text{im}} = 1 - \frac{F\omega^2}{\omega^2 - \frac{1}{CL} + i\frac{Z(\omega)\omega}{L}}, \quad (1)$$

where  $F$  is the filling factor,  $C$  and  $L$  are the geometrical capacitance and inductance, and  $Z(\omega)$  is the ring metal impedance. Using equation (1) we calculated the frequency dispersion of  $\mu_{\text{eff}}$  from 100THz to 800THz covering the entire visible light frequency region taking into account the electro-magnetic properties of the metals (silver, gold, and copper).

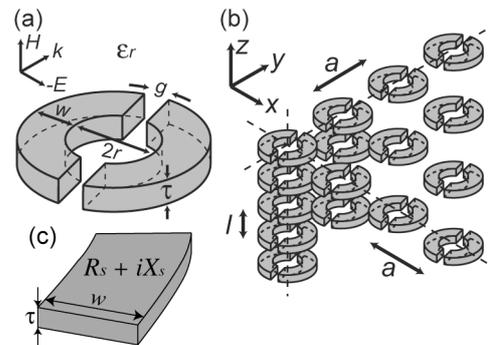


Figure 2 Models of metamaterial structure used in calculations.

As shown in Figure 3(a), the effective permeability  $\mu_{\text{eff}}$  changes both positively and negatively at the resonant frequency of the SRRs array. In Figure 3(b), we plotted the calculation results of the change of  $\mu_{\text{eff}}$ , which means the difference between  $\max \mu_{\text{eff}}$  and  $\min \mu_{\text{eff}}$  as shown in Fig. 3(a), for each metal SRRs array. From these results we have

clarified that a three-dimensional array of split-ring-resonators made of silver can give a strong magnetic response in the visible light frequency region. As also shown in Fig. 3(b), silver SRRs exhibit  $\mu_{\text{eff}}$  changes exceeding 2.0 in the entire visible range, which means  $\mu_{\text{eff}}$  can become a negative value, while the responses of gold and copper SRRs do not exceed 2.0 in the visible light region.

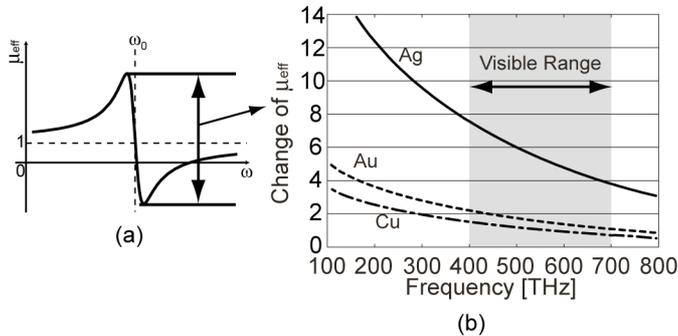


Figure 3 Frequency dependencies of the change of effective permeability ( $\mu_{\text{eff}}$ ) of SRRs made of silver, gold, and copper.

### III. TWO-PHOTON REDUCTION METHOD

To create a plasmonic metamaterial structure, the fabrication technique requires the ability to make arbitrary three-dimensional metallic structures. To satisfy this requirement, we have proposed a new fabrication technique that uses two-photon induced reduction of metal complex ions [5-9].

Figure 4 shows a schematic of the two-photon reduction technique. A mode-locked Ti:Sapphire laser system with a center wavelength of 800 nm, a pulse width of 80 fs, and a repetition frequency of 82 MHz was used as a light source. The laser beam was focused inside the metal-ion solution using an oil-immersion objective lens. The focused laser beam was scanned in two dimensions (x-y scanning) using a pair of galvanometer mirrors, and was also scanned in the longitudinal direction (z-scanning) by translating the objective lens using a computer-controlled motor stage.

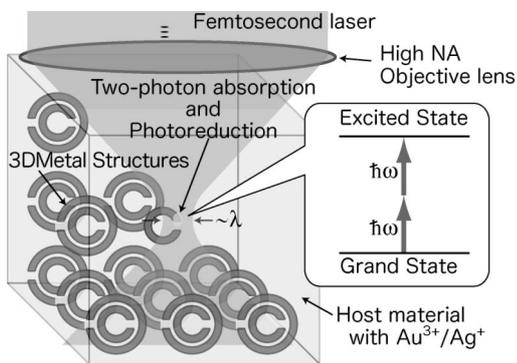


Figure 4 Schematic of two-photon-induced metal-ion reduction by focused fsec NIR laser irradiation.

Figure 5 shows the 3D metal structures fabricated using a silver ion aqueous solution with Coumarin 440 dye. Fig. 5(a) and (b) show a silver 3D gate structure and a top-heavy silver cup, respectively. Fig. 5(c) shows 3D silver pyramid structures with 200nm line width. This nano-meter scale resolution was achieved by the metal crystallization control using dopant surfactant molecules. These structures also have sufficient strength to endure cleaning with water; that is, they were not destroyed by the surface tension of the meniscus when removing the ion solution. The resistance of the 3D silver gate (Fig. 5(a)) was 4.59 k $\Omega$  as measured by connecting each foot to a silver electrode with silver wire. This result indicates that the fabricated 3D silver microstructure is also electrically conductive.

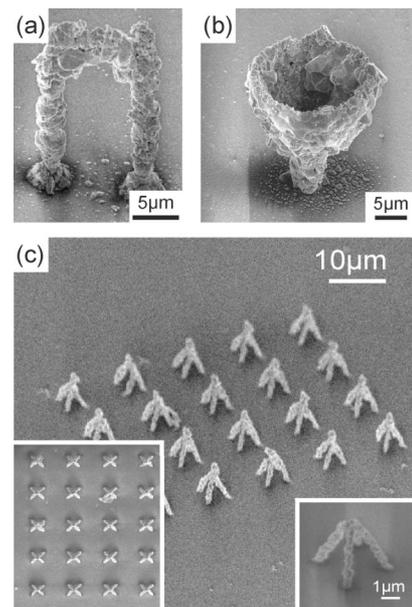


Figure 5 3D metal structures fabricated by two-photon reduction technique.

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