Femtosecond laser-induced forward transfer for multilayer plasmonic metamaterials

Pin Chieh Wu, Ming Lun Tseng, Wei Ting Chen, Chun Yen Liao, Shulin Sun, Chia Min Chang, Cheng Hung Chu, Po-Li Chen, Lei Zhou, Ding-Wei Huang, Ta-Jen Yen, and Din Ping Tsai

Applying a laser-direct-writing technique to the fabrication of split-ring-resonator-based metamaterials offers a straightforward alternative to complicated, time-consuming conventional approaches.

Metamaterials are artificially engineered composites that exhibit extraordinary optical properties not found in nature, such as negative refraction. These materials have potential application in ultra-high-resolution imaging devices and nanolasers based on plasmonics. So-called plasmonic metamaterials exploit surface plasmons, a form of surface electromagnetic radiation triggered by light. The split-ring resonator (SRR) is one of the most commonly used building blocks for constructing plasmonic metamaterials. These U-shaped structures are individually designed to produce a particular magnetic response—known as plasmonic resonance—to the electromagnetic field. When incident light hits the plasmonic resonance of SRR-based metamaterials, the behavior of the composite changes, depending on the nature of the resonance. In addition, a near-field coupling effect between different plasmonic metamaterials gives rise to unusual physical phenomena, such as Fano resonance and a toroidal dipolar response. These effects are particularly interesting for the development of sensors and slow-light devices, to give just two examples.

Ideas and strategies for practical application abound in the field of plasmonic metamaterials. However, several problems must first be overcome. One challenge is that to qualify as a real material, metamaterials must comprise truly 3D multilayer unit cells. Technologies used to date to fabricate multilayer and 3D plasmonic structures include electron beam lithography, in which alignment errors caused by the multiexposure process make fine control very difficult.

Here, we describe work that uses contact-mode femtosecond laser-induced forward-transfer (fs-LIFT), a laser-direct-writing (LDW) approach to making multilayer metamaterials. In contact-mode fs-LIFT, a ‘thrust’ or transfer film absorbs energy from focused fs-laser pulses, which dramatically increases pressure at the interface between the film and the ‘donor’ substrate. When the temperature of the thrust material and overlying films rises above the melting point, they are locally transferred forward to a ‘receiver’ substrate. This process leaves behind uniform, multilayer-structured metamolecules on the donor substrate.

We sputtered successive multilayer films of germanium antimony tellurium (Ge$_2$Sb$_2$Te$_5$, 10nm), gold (Au, 20nm), zinc sulfide–silicon dioxide (ZnS–SiO$_2$, 50nm), and Au (20nm) onto

Figure 1. (a) Schematic showing fabrication of multilayer plasmonic structures by contact-mode femtosecond laser-induced forward-transfer (fs-LIFT). (b) Separating the laser-processed donor-receiver pair yields multilayer plasmonic structures. (c) Feature dimensions of the multilayer metamaterial. BK-7: Glass. as-Ge$_2$Sb$_2$Te$_5$: As-deposited germanium antimony tellurium. Au: Gold. ZnS-SiO$_2$: Zinc sulfide–silicon dioxide.

Continued on next page
transparent BK7 glass (the donor). Another glass substrate in close contact with the surface of donor acts as the receiver. The Ge$_2$Sb$_2$Te$_5$ thin film acts as the thrust material for fully transferring the overlying films from donor to receiver.

Figure 2(a, b) shows scanning electron microscopy images of fabricated SRR arrays on the donor and the corresponding receiver. The fluence of the incident laser beam is maintained at 306 mJ/cm$^2$, and the raster speed at 53.3 μm/s. Following fs-LIFT, we observed multilayer SRRs whose individual shapes were clear and sharp (see Figure 2). The insets of Figure 2(a, b) show the zoomed images of individual SRRs on the donor and receiver.

Figure 3(a, b) shows the experimental and simulated reflection spectra of the sample, respectively. The insets indicate the configuration of the light excitation. Our experimental results are in good agreement with simulations both for x-polarized—see Figure 3(a)—and y-polarized illumination: see Figure 3(b). In the case of x-polarization, three pronounced resonance peaks appear at 21.0, 30.9, and 52.7 THz, respectively. In contrast, for y-polarized illumination we observe only one resonance peak at a frequency around 29.6 THz. We believe the slight quantitative differences between the experimental and simulation results may be caused by imperfections in fabricating real samples compared with theoretical designs, as well as inaccuracies in the dielectric constants of real materials adopted in finite-difference-time-domain (FDTD) simulations.

To better understand the nature of the plasmonic resonance peaks, we simulated and plotted the induced-current distributions at points of resonance on the surfaces of structures under normally incident light with x- and y-polarization, respectively. The first (second) row of Figure 4 shows the current density distributions on the upper (lower) Au layer. As we expected, under normal illumination, the magnetic resonances of these planar SRR structures can only be excited by the x-polarized wave. Most interestingly, the currents on two prongs of the U-shape are in phase, while those on the bottom rods of two SRRs are out of phase, due to different near-field coupling strengths in different regions of the Au SRR. The charges with anti-phase oscillation are generated on the surface of the lower SRR structure when the induced electric dipole of the upper Au SRR couples to the lower Au SRR. Comparing the induced currents in the upper and lower SRRs, we found that they are significantly different only on the bottom rod. On the two prongs, they are nearly identical. This peculiar behavior implies that the near-field coupling between two Au layers is most significant in the two bottom rods.

In the case of y-polarization, both the upper and lower Au layers show an antiphase electric resonance coupling that results in a pronounced resonance peak at 29.6 THz. In principle, each single-SRR mode should split into a mode pair in the double-SRR system due to the coupling effect. In general, there should be a coupling effect between two plasmonic metamaterials in the near-field region. However, in our results, there is only one pronounced resonance peak with multilayer structures under y-polarized illumination. We conclude that the coupling effect between plasmonic metamaterials is weaker when the distance between them is much shorter than the wavelength of incident light.

In summary, we successfully fabricated multilayer SRRs by applying contact-mode fs-LIFT. Our results show that application of the proper laser fluence to the multilayer films is important in ensuring that samples are uniform and smooth. We analyzed reflection spectra of a multilayer SRR array and found them to be in excellent agreement with FDTD simulations, both exhibiting rich resonance properties. We investigated both...

Continued on next page
Figure 4. Simulation current density at plasmonic resonant frequencies for (a) x-polarized and (b) y-polarized illumination. Insets: The designed structure of the unit cells.

electric and magnetic resonances by calculating the induced current patterns. This work points to the promise of fs-LIFT for straightforward fabrication of multilayer micro- and nanostructures for practical applications such as plasmonic devices and 3D metamaterials. We expect that using substrates such as flexible, transparent sheets and optical fibers will increase the utility of fs-LIFT. Based on these results, our future work will focus on achieving larger chiral (left- or right-handed) optical effects in multilayer structures due to their greater sensitivity to magnetic fields.

Author Information

Pin Chieh Wu and Ming Lun Tseng
Graduate Institute of Applied Physics
National Taiwan University
Taipei, Taiwan
and
Instrument Technology Research Center
National Applied Research Laboratories
Hsinchu, Taiwan

Wei Ting Chen
Graduate Institute of Applied Physics
National Taiwan University
Taipei, Taiwan

Chun Yen Liao and Cheng Hung Chu
Department of Physics
National Taiwan University
Taipei, Taiwan

Shulin Sun
Department of Physics
and
National Center of Theoretical Sciences at Taipei
Physics Division
National Taiwan University
Taipei, Taiwan

Chia Min Chang and Ding-Wei Huang
Graduate Institute of Photonics and Optoelectronics
National Taiwan University
Taipei, Taiwan

Po-Li Chen
Instrument Technology Research Center
National Applied Research Laboratories
Hsinchu, Taiwan

Lei Zhou
State Key Laboratory of Surface Physics
and
Key Laboratory of Micro and Nano Photonic Structures
Fudan University
Shanghai, Taiwan

Ta-Jen Yen
Materials Science and Engineering
National Tsing Hua University
Hsinchu, Taiwan

Continued on next page
Din Ping Tsai  
Graduate Institute of Applied Physics  
National Taiwan University  
Taipei, Taiwan  
and  
Research Center for Applied Sciences  
Academia Sinica  
Taipei, Taiwan  

References  