Terahertz metamaterials fabricated by inkjet printing

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Metamaterial layers designed for gigahertz to terahertz (THz)-frequencies have been fabricated by inkjet printing. The spectral response of the structures consisting of periodically arranged metallic split-ring resonators is characterized by THz-time-domain spectroscopy and compared with identical structures produced by conventional photolithography and etching techniques. The broader linewidth of their resonances is shown to originate mainly from structural inhomogeneities. Our study shows that inkjet printing is a viable route for producing metamaterial structures, allowing for rapid processing and flexibility in the choice of substrates. © *2009 American Institute of Physics*. [doi:10.1063/1.3276544]

Metamaterials have attracted considerable interest in recent years, mainly due to the range of exotic applications they make possible, which are not attainable by naturally occurring materials.[1–3] Since their first experimental demonstration in the microwave regime, progress in structure miniaturization extended the spectral range for metamaterial applications over terahertz and infrared to optical frequencies.[4,5] Located at the interface between electronic and optical regimes, the terahertz (THz) range is particularly suited for the detailed investigation of metamaterial properties. Due to the moderate structure sizes at THz wavelengths and the availability of powerful characterization techniques, such as broadband and phase-resolved spectroscopy[6] or near-field microscopy,[7] the THz regime provides the ideal testbed for metamaterial designs. The inherent scalability of metamaterial structure allows the transfer of concepts to other frequencies. Moreover, the use of metamaterial surfaces for biological and chemical sensing applications has been demonstrated at THz frequencies, where many compounds show characteristic absorptions.[8–10]

Currently, the production of THz metamaterials is mainly based on standard microfabrication techniques, requiring clean-room technology with the corresponding photolithography, and metal deposition or etching steps. Recent reviews on inkjet printing[11] have discussed its appeal as a versatile and low-cost manufacturing technique, and its employment for a number of applications beyond its widely known use in graphics. Examples of such areas are organic electronics,[12] tissue engineering,[13] and thin film transistor liquid crystal display color filters.[14] The attraction of inkjet printing lies in its ability to precisely deposit picoliter sized droplets at predetermined positions on a substrate using computer-controlled translation stages and ink dispensers. This ability removes the need for masks, which leads to a reduction in the number of necessary process steps. This reduction, in turn, enables cost-savings and the elimination of waste to be made, as well as allowing a highly efficient use of materials. Moreover, contamination is minimized since inkjet printing is a noncontact deposition process.

In this letter we demonstrate the fabrication of split-ring resonator (SRR) arrays designed for gigahertz to THz frequencies by inkjet printing. Their resonant behavior is investigated by terahertz time-domain spectroscopy (THz-TDS) and is compared with structures produced by conventional microfabrication techniques.

Figure 1 shows different SRR structures fabricated by inkjet printing. The ink used for the printing was a 20 wt % suspension of silver nanoparticles, whose sizes ranged between 20–50 nm, dispersed in ethanol and ethylene glycol (U5603, Sun Chemicals, Slough, GB). The substrate was 125 μm thick polyimide (Kapton®, 500HN, Krempel, Vaihingen/Enz, Germany). The inkjet printing was performed using the Dimatix DMP 2831 (Dimatix-Fujifilm Inc., Santa Clara, USA), which was equipped with a piezoelectric printhead cartridge (DMCLCP-11610) that dispenses droplets with a nominal volume of 10 pL. Only one nozzle was used for the printed structures that are reported here. The printing frequency was set at 5.0 kHz, and a customized waveform[15] was used. The temperature of the platen and the print cartridge was set to 55 °C. Two samples have been

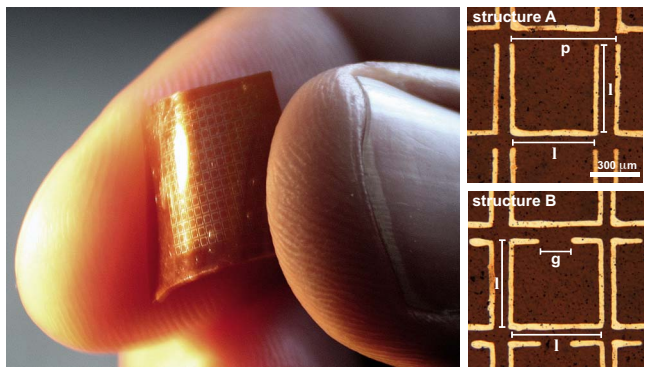

FIG. 1. (Color online) Inkjet printed SRR arrays on a flexible polyimide substrate. Two different structures have been fabricated (l=500 μm, p=600 μm, g=200 μm).

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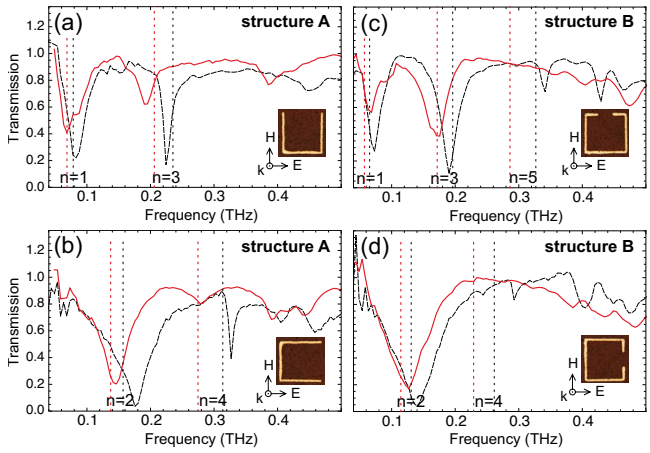

FIG. 2. (Color online) (a) and (b) show transmission spectra of square arrays of structure A fabricated by inkjet printing (solid red curve) and by conventional microfabrication (dashed black curve) for two perpendicular orientations of the THz electric field as indicated in the insets. (c) and (d) show corresponding transmission spectra of arrays of structure B. The vertical dotted lines in all spectra indicate the eigenmode frequencies calculated according to Eq. (1).

printed, one consisting of a 10×10 square array of U-shaped SRRs (structure A), and a second one with additional arms forming small gaps (structure B). The as-printed samples were converted to conductive silver by being placed for 150 min on a hot-plate that was set to 220 °C. This duration and temperature were used in order to obtain the highest possible conductivity ($\sim0.3-0.5\times$bulk silver).

Reference structures with identical geometry were produced by conventional photolithography and etching techniques from 9 μm thick copper on a 120 μm thick polytetrafluoroethene (PTFE) substrate from Rogers (RT/duroid 5880®). First, a layer of photoresist was spin-coated on the surface. After UV-light exposure through a photomask, the photoresist layer was subjected to development which destroys unwanted areas of the protective layer, exposing the corresponding areas of the copper. Subsequent chemical etching removes the exposed metal from the substrate.

All samples were characterized by conventional THz-TDS based on the coherent emission and detection of free-space THz pulses from optically gated photoconductive antennas.[6] The relative amplitude transmission through the structures was determined from measurements of the terahertz electric field transmitted through the SRR sample and through the bare substrate as a reference.

Transmission spectra of the inkjet printed structures together with spectra of the etched references are shown in Fig. 2. Measurements were made with the sample plane normal to the beam for two orthogonal polarizations of the electric field relative to the SRR structure, as shown in the insets. For discrete frequencies the incident electromagnetic field is able to excite current density standing waves on the SRRs.[16,17] These resonances occur roughly at frequencies where

$$L = n\lambda_m/2, \tag{1}$$

where L is the length of the unfolded SRR and λ_m the wavelength in the surrounding medium. n is an integer number counting the resonance order. When the electric field is parallel to the open side of structure A [Fig. 2(a)] two dominant transmission minima below 0.3 THz are observed for the inkjet printed and the etched sample. In this configuration only odd eigenmodes are excited ($n=1,3,5\ldots$), i.e., where the current density standing wave has an even number of nodes along the metallic structure. The vertical dotted lines indicate the estimated frequencies of the two lowest-order eigenmodes $n=1$ and 3 for both samples calculated by taking the different dielectric environment into account. For simplicity we assumed a homogeneous dielectric background with an effective permittivity given by the average of air and the substrate, with $\varepsilon=1.8$ for the kapton and $\varepsilon=1.5$ for the PTFE substrate, respectively. The substrate permittivities have been determined from separate THz-TDS measurements. For orthogonal polarization only even eigenmodes can be excited. As shown in Fig. 2(b), again the two modes at lowest frequency ($n=2$ and 4) are observed. Note that in all spectra additional transmission minima occur above 0.38 THz, which are almost independent on polarization. Here the periodicity of the lattice leads to abrupt changes in transmission due to the so-called Wood anomalies,[18] which occur whenever a new grating order changes from evanescent into propagating. Figures 2(c) and 2(d) show the transmission spectra of an inkjet printed and an etched version of structure B. Also in this case, the lowest order eigenmodes are observed for both sample types and terahertz polarizations.

Our main observation in all spectra is that the transmission minima associated with the structural eigenmodes occur (i) weaker, (ii) significantly broadened, and (iii) shifted to lower frequencies for the inkjet printed samples relative to the etched reference arrays. As indicated by the vertical dotted lines in Figs. 2 the spectral shifts can be attributed to the different substrate permittivities. On the other hand, the observed line-broadening and the shallower absorption minima of the inkjet printed structures can in principle originate from different mechanisms. Generally, a lower metal conductivity has an effect on the transmission amplitude and the linewidth due to enhanced ohmic damping of the resonant currents in the metal. However, in the THz regime this effect is typically weak, owing to the rather large conductivity of even poor metals so that radiation damping is the dominant damping mechanism.[19]

Microscopic inspection of our structures reveals geometric variations of the inkjet printed SRR arrays. Apart from fluctuations of the line width of the SRRs we observe significant variations in the periodicity p and the side lengths l. Simple disorder in arrays as introduced by randomly varying periodicities in principle leads to shallower transmission minima but cannot account for the observed line-broadening.[20] A variation in side lengths, however, introduces inhomogeneous broadening to the transmission spectra. Figures 3(a) and 3(b) show histograms of the side length variations (l in Fig. 1) for structure A both, fabricated by etching and by inkjet printing. Both side length distributions are centered close to 500 μm, however, whereas for the etched sample the length deviates from its mean value by $<2\%$, the size of the inkjet printed structures shows a variation on the order of $>10\%$, which corresponds to approximately ±25 μm, the diameter of one ejected droplet. The length variations can be translated into a spread of eigenfrequencies according to Eq. (1). As an example, we plot the resulting histograms of the mode frequencies calculated for the $n=3$ mode together with the measured extinction spectra in Fig. 3(c). Since for a SRR the resonances are typically slightly shifted to lower frequencies compared to a wire of

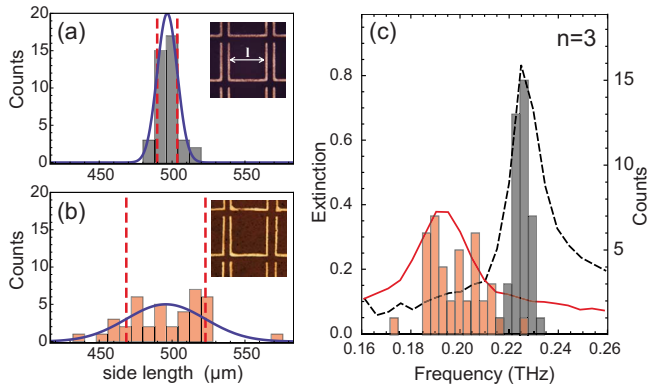

FIG. 3. (Color online) Histograms showing the side length distribution for the etched (a) and the inkjet printed (b) SRR arrays (structure A). The insets show photographs of the structures. (c) Corresponding histograms of calculated resonance frequencies for the $n=3$ mode using Eq. (1), shown together with the measured extinction spectra.

same length owing to the different geometric charge distribution,[21] we introduced a relative redshift of 5% to the calculated frequencies. The match between the spectral linewidths and the width of the predicted frequency distributions indicates that the variation in structure size can indeed account for the observed line broadening in the spectra. Future advances in optimizing the printing process to minimize structural disorder and size variations would therefore permit the production of metamaterial structures with a spectral response comparable to conventionally manufactured structures. This may be achieved by using a different substrate since polyimide becomes rougher after heat treatment. The sintering time and temperature can be reduced if higher conductivities are not essential. A recently developed room temperature process which takes 5 min and uses PET as a substrate may be an appealing solution.[22]

In conclusion our results demonstrate that inkjet printing, which has the advantages of being a fast, flexible, and low-cost alternative to conventional microfabrication techniques, can be considered a method for manufacturing THz metamaterials. However, in order to reduce variations, such as reported in this letter and may be considered a hindrance, a reduction in droplet size may be desirable.

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