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Fabrication of Terahertz Planar Metamaterials Using a Super-Fine Ink-Jet Printer

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Super-fine ink-jet (SIJ) printing technology is applied to the fabrication of terahertz metamaterials. A silver film is fabricated using an SIJ printer with silver paste ink, and it is confirmed that the film behaves as a good conductor in the terahertz frequency region. Then, basic terahertz metamaterials such as metal wire-grid structures and split-ring resonators are printed on high-resistivity silicon substrates. The terahertz responses of the printed samples agree with those expected from their structures. SIJ printing is one of the ideal methods for fabricating terahertz metamaterials owing to its rapidity, simplicity, flexibility, and sufficient accuracy.

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In the terahertz frequency region, novel devices based on metamaterial concepts have been developed, examples of which include terahertz wave amplitude modulators, phase modulators, and absorbers.[1–3] These terahertz devices are designed with various kinds of planar metallic patterns. The diversity of the metallic patterns contributes to the construction of various advanced terahertz systems and their applications.[4] Photolithography is one of the powerful methods for fabricating highly accurate terahertz planar metamaterials composed of metallic lines having widths of a few microns. However, photolithography involves rather complicated processes such as preparing photomasks even for a few samples.

Recently, an alternative method — an ink-jet printing method — has become available for such fabrication. Murata *et al.* have developed a super-fine ink-jet (SIJ) printer for nanotechnology research and application to microelectronics.[5–8] In SIJ printing, only printing and annealing processes are carried out to fabricate two-dimensional metallic patterns with accuracy comparable to that of photolithography. Using the SIJ printer, one can print arbitrary metallic patterns on various kinds of substrates (semiconductor, glass, polymer, and other smooth substrates). The SIJ printing process is operated in ordinary atmosphere and does not require special clean rooms. Additionally, it is also noteworthy that the SIJ printer has the ability to fabricate three-dimensional structures.[6,8] The SIJ printing process is far simpler and more flexible than the photolithography process.

In this paper, we propose the use of the SIJ printer for the fabrication of terahertz metamaterials. Wire-grid (WG) structures and split-ring resonators (SRRs) are fabricated using the SIJ printer. For printing metallic patterns, gold and silver pastes (Nanopaste™)[9,10] are used as the ink. Nanopaste is composed of single-dispersion metallic nanoparticles, whose diameter is approximately 5 nm, and the minimum achieved diameter of a droplet is of the order of submicrons. For the metallization of the silver paste, a heat-treatment process must be carried out at 220 °C for 1 h for sintering. After sintering, the dc resistivity of the silver paste becomes 3 μΩ cm,[6] which is slightly larger than the resistivity of bulk Ag, 1.61 μΩ cm.[11] For using this Nanopaste as a material for metamaterials in the terahertz region, it is necessary to confirm that it has sufficient conductivity in this frequency region.

First, we fabricated a silver film on a high-resistivity silicon (Si) substrate (refractive index $n_s = 3.42$ and thickness $d_s = 0.48$ mm) with the SIJ printer in order to evaluate the terahertz response of silver Nanopaste. After printing, the film and substrate were heated at 220 °C for 1 h. From the atomic force microscopy (AFM) image of the film, its thickness d_f and its root mean square (RMS) were estimated to be approximately 240 and 82 nm, respectively. The complex permittivity of the film was measured using a terahertz time-domain spectroscopy (THz-TDS) system.[12,13] The relative permittivity of the silver film is fitted by the Drude model $\varepsilon = 1 - \omega_p^2\omega^{-1}(\omega + i\tau^{-1})^{-1}$. Here, ω is the angular frequency, $\omega_p = 2\pi f_p$ is the angular plasma frequency, and τ is the relaxation time. f_p and τ are estimated to be 320 THz and 0.19 ps, respectively. In ref. 14, the relative permittivity of bulk silver was reported to be $\varepsilon = -193000 + i731000$ at 1.2 THz.[14] The annealed silver film shows $\varepsilon = -47700 + i33300$ at 1.2 THz, which is smaller than that in ref. 14. However, the relative permittivity of the silver film is comparable to that of bulk gold in the terahertz region (e.g., in ref. 14, for gold, $\varepsilon = -28000 + i277000$ at 1.2 THz). The skin depth of the printed silver, $\delta = \sqrt{2/(\mu_0\sigma_{dc}\omega)}$, is estimated to be 193 nm at 1 THz. Here, μ_0 is the permeability of vacuum and $\sigma_{dc} = \varepsilon_0\omega_p^2\tau \sim 68000$ S/cm is the dc conductivity. Singh *et al.* evaluated the terahertz responses of the SRR array as a function of metal thickness.[15] They indicated that metal structures thicker than the skin depth were required to saturate the transmission minimum due to *LC* resonances. When fine lines with a width of a few microns are printed by the SIJ printer, the thickness of the lines becomes 100–200 nm. This thickness is comparable to the skin depth and the thickness of the metallic patterns fabricated by the conventional photolithography process. Additionally, metal lines several microns thick are printed by simply iterating the printing process. Thus, SIJ printing using the silver Nanopaste ink can be one of the ideal methods for fabricating terahertz metamaterials.

Metal WG structures have been used as polarizers and high-pass filters in the terahertz frequency region.[16] In recent years, WG structures are considered as the metama-

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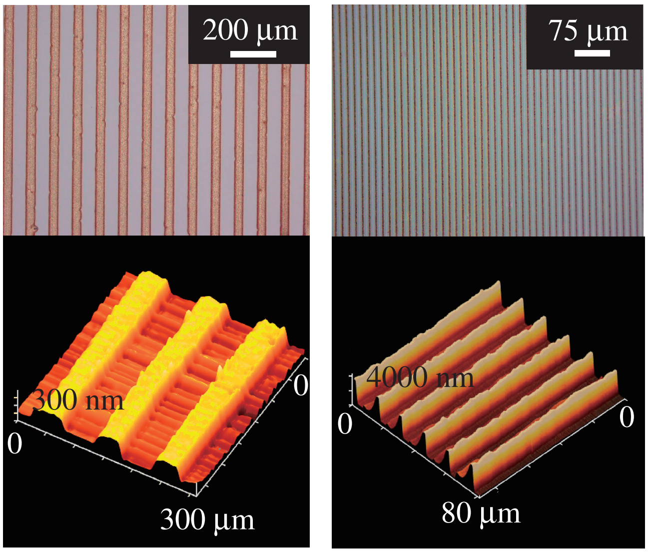

(a) (b)

Fig. 1. Microscope photograph and AFM images of wire-grid patterns printed on Si substrates. The periods of the wires are (a) 100 μm (pattern A) and (b) 15 μm (pattern B).

terials with the negative permittivity.[17)] Two patterns of WG structures were printed using the SIJ printer (Fig. 1). Patterns A and B have line periods p of 100 and 15 μm, respectively. Figure 1 shows the optical microscope photographs and AFM images of the WG patterns printed with the silver paste on high-resistivity Si substrates. Pattern A has 40-μm-wide lines printed by drawing approximately 20-μm-width lines displaced three times by 10 μm successively. From the AFM image, the thickness is estimated to be 240 nm, which is thicker than the skin depth. Pattern B has 5-μm-width fine lines with a thickness of 3000 nm. This thickness was achieved by drawing ten times without displacements and is far greater than the skin depth. As demonstrated here for the two WG patterns, the thickness and width of the metallic lines can be controlled by the SIJ printer without requiring any complicated processes. Such control is useful for the rapid optimization of the structures of the terahertz devices.

Figure 2 shows the transmission spectra of the WG patterns normalized by that of the Si substrates. For transverse magnetic (TM) waves (electric fields perpendicular to the wires), the transmittance of pattern A decreases with frequency above the first-order diffraction frequency $f_\mathrm{d} = c/n_\mathrm{eff}\,p \sim 0.88$ THz (open circles in Fig. 2). Here, we use the effective refractive index around the lines, $n_\mathrm{eff} \sim n_\mathrm{s}$. For transverse electric (TE) waves (electric fields parallel to the wires), the transmission spectrum of pattern A shows a broad peak at 0.90 THz, which is near f_d (open squares in Fig. 2). For the TE waves, the electromagnetic waves have a cutoff frequency of $f_\mathrm{c} = c/2n_\mathrm{eff}w$ if we consider the gaps between metallic lines as waveguides, with w as the line spacing. The TE waves propagate between the metallic lines at frequencies above $f_\mathrm{c} \sim 0.73$ THz. The broad peak at 0.90 THz of pattern A is attributed to a waveguide resonance of the TE waves via diffraction.[18,19)] This property allows the pattern to be used as a high-pass filter. The transmittance of pattern B is greater than 0.9 below 2 THz for TM waves because a cutoff frequency does not exist for the TM waves (closed circles in Fig. 2). In contrast, the transmittance of pattern B is less than 0.1 for TE waves (closed squares in Fig. 2). Because the propagation constant β is an imaginary number for frequencies lower than f_c, the transmittance exponentially decreases with metal thickness. Here, f_c is estimated to be 4.4 THz for pattern B by using $n_\mathrm{eff} \sim n_\mathrm{s}$. This property allows the pattern to be used as a polarizer in the terahertz frequency region.

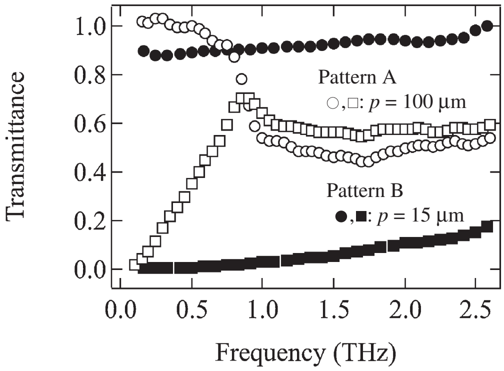

Fig. 2. Transmission spectra of wire-grid patterns printed on Si substrates. The incident terahertz wave polarizations are perpendicular (circles) and parallel (squares) to the wires.

The second metamaterial pattern fabricated using the SIJ printer was the SRR. SRRs are the most commonly used metallic structures for designing the magnetic responses (permeability) of metamaterials.[20)] Additionally, closed-ring resonators (CRRs) were fabricated for comparison. Figures 3(a) and 3(b) show optical microscopy photographs of the SRRs and CRRs, respectively, printed on Si substrates with silver paste. Figure 3(c) shows a topographic image of the SRR measured with a laser scanning microscope. The $5 \times 5\,\mathrm{mm}^2$ samples comprised 2500 ring elements. The split gap width, line width, side length of the ring, and ring period were 8, 4, 42, and 100 μm, respectively. The printing process was iterated three times to make the lines thicker than the skin depth and required about 10 h for drawing. As a result, the thickness became approximately 300 nm, as measured with the laser scanning microscope.

Measured transmission spectra of the SRRs and CRRs are shown in Figs. 3(d) and 3(e), respectively. For the incident polarization parallel to the SRR gap [polarization A in Fig. 3(a)], a transmission dip is observed at 0.8 THz [solid red line in Fig. 3(d)]. This dip is attributed to the half-wave resonance of a pair of sidelines of the ring.[21)] The transmission spectra of the CRRs also show a dip at 0.8 THz, which is attributed to the half-wave resonance for both polarizations A and B [solid red and blue lines in Fig. 3(e)]. This result implies that the existence of the gap of the SRR does not affect the dip frequencies for the polarization A. For the incident polarization perpendicular to the SRR gap [polarization B in Fig. 3(a)], the transmission spectrum of the SRRs shows two dips at 0.4 and 1.1 THz [solid blue line in Fig. 3(d)]. The resonance at 1.1 THz is attributed to the half-wave resonance of a single sideline of the ring. The transmission dip of the SRRs at 0.4 THz is induced by the LC resonance associated with circulating currents.[21)] The LC resonance at 0.4 THz vanishes in the CRRs because the

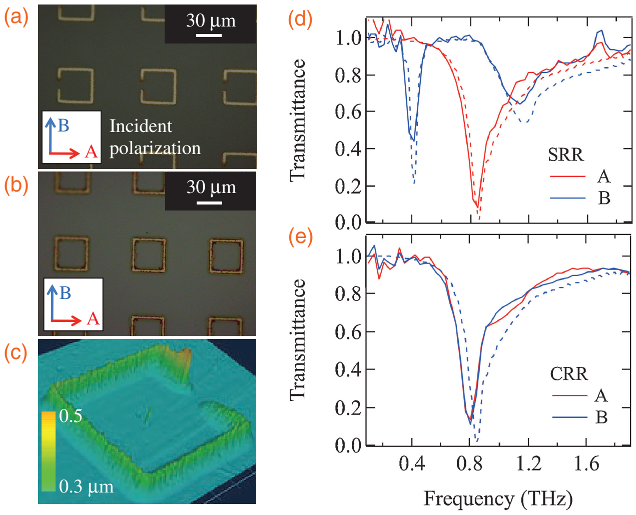

Fig. 3. Microscope photographs of (a) SRRs and (b) CRRs printed on Si substrates, and (c) topographic image of SRRs. Transmission spectra of (d) SRRs and (e) CRRs for incident polarizations A (red) and B (blue). The dashed lines show the transmittance simulated using the finite-difference time-domain method.[24]

effective capacitance becomes infinite, causing the resonant frequency of the LC circuit to become zero. This LC resonance can be controlled by varying the conductivity of the substrate in the SRR gaps with optical excitations or bias voltages.[1,2]

Finally, we point out that the SIJ printing technology is useful in the application of SRRs to sensing permittivity of small volume materials. The use of SRRs for sensing is proposed because the frequency of the LC resonance is sensitive to the permittivity of the materials near the SRR gap.[22] The sensitivity increases with increasing metal thickness.[23] As demonstrated for the WG structures in this study, metal structures with a thickness of several microns can be fabricated using the SIJ printer by simply iterating the printing process.

In this study, we used the SIJ printer for fabricating terahertz metamaterials. We fabricated a silver film using the SIJ printer with silver Nanopaste to evaluate its terahertz responses. The permittivity of the silver film is comparable to that of bulk gold in the terahertz frequency region. The patterns printed with the silver paste can be thicker enough than the skin depth in order to fabricate terahertz metamaterials. The basic metamaterials, i.e., the WG structures and SRRs, were fabricated using the SIJ printer; their electromagnetic responses agree with those expected from their structures. The greatest advantage of the SIJ printing method over other highly accurate fabrication methods is that arbitrary metal patterns with various line widths and thicknesses can be fabricated without involving any complicated processes. SIJ printing is one of the ideal methods for fabricating terahertz metamaterials owing to its rapidity, simplicity, flexibility, and sufficient accuracy. SIJ printing has the potential to accelerate the development of the terahertz metamaterials.

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