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FAST TRACK COMMUNICATION

# Terahertz metamaterials on free-standing highly-flexible polyimide substrates

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**Abstract**

We have fabricated resonant terahertz metamaterials on free-standing polyimide substrates. The low-loss polyimide substrates can be as thin as 5.5 $\mu$m yielding robust large-area metamaterials which are easily wrapped into cylinders with a radius of a few millimeters. Our results provide a path forward for creating multi-layer non-planar metamaterials at terahertz frequencies.

(Some figures in this article are in colour only in the electronic version)

The advent of metamaterial composites has given rise to numerous electromagnetic functionalities previously unimagined. This includes negative refractive index, superlensing, cloaking and quite generally, the fabrication of metamaterials which have been designed using coordinate transformation approaches [1–5]. Many of these ideas were initially implemented at microwave frequencies where fabrication of multilayer composites has become increasingly sophisticated during the past several years. This has resulted in dramatically reduced times from conceptualization and electromagnetic simulation to, ultimately, fabrication and characterization. However, the fabrication of subwavelength unit cells becomes increasingly challenging in moving from the microwave to visible region of the electromagnetic spectrum though important progress has been made [6–9]. To date, the majority of this work has been on planar composites. At terahertz (THz) frequencies and above, creating multiple unit cell structures in the direction of propagation and taking full advantage of coordinate transformation MM design to realize non-planar MM composites requires the development of new fabrication strategies.

The far-infrared, or terahertz, is a promising region to investigate novel approaches to metamaterials fabrication. First, the unit cells are on the order of tens of micrometres which is amenable to novel microfabrication approaches. Second, and perhaps of greater importance, there is a strong technological impetus to create sources, detectors and components at terahertz frequencies to realize the unique potential of THz radiation [10–13]. Metamaterials are expected to play an important role in this regard as evidenced by recent demonstrations of MM-based modulators and frequency tunable filters [14–16]. An important step in the progression of functional THz MM composites is the fabrication of multilayer structures. For example, a strongly resonant THz MM absorber consisting of two layers spaced by approximately 6 $\mu$m [24] was recently demonstrated (see also [17] for the microwave perfect absorber). The entire structure, however, was on top of a thick GaAs supporting substrate. There has also been other work at THz frequencies using polymer spin-coating based techniques to fabricate metamaterials, but the focus of this work was not on ultrathin flexible substrates [18, 19].

Closely related to creating THz metamaterial composites is the seminal work on frequency selective surfaces [20–22]. More recently, there has been a report on creating polyimide-based multilayer metallic photonic band-gap (MPBG) structures at terahertz frequencies [23]. The MPBG structure

 

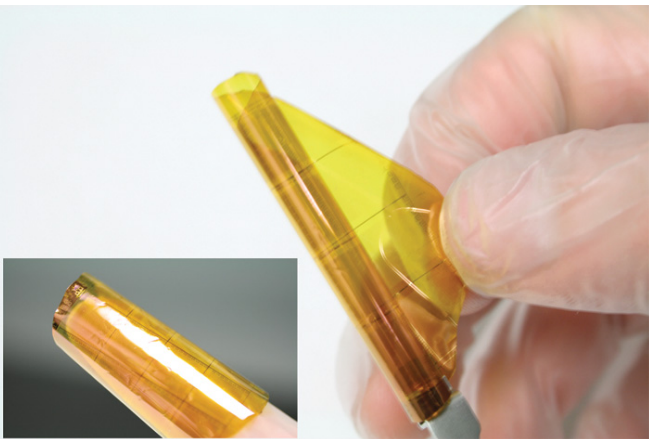

**Figure 1.** Photographs of the free-standing electric metamaterials fabricated on polyimide substrate. The thin samples naturally roll up into a cylinder unless supported on a frame. (Inset) A flexible 'skin' applied to a finger. (Colour online.)

served as a −35 dB notch filter at ∼4.5 THz. The primary difference between [23] and the present publication is that (a) we are focusing on split ring resonators and (b) our structures are easily peeled from the substrate while in [23], a citric acid/hydrogen peroxide etch was used to remove the substrate.

It is important to extend such work and develop MM on thin flexible substrates that are considerably thinner than the lateral unit cell dimensions in moving towards creating multilayer non-planar metamaterials such as cloaks, concentrators or resonant absorbers at terahertz frequencies. In this letter, we demonstrate resonant terahertz metamaterials on free-standing low-loss polyimide substrates as thin as 5.5 $\mu$m yielding robust large-area metamaterials which are easily wrapped into cylinders with a radius of a few millimetres.

The MM structures were fabricated by depositing a 200-nm-thick gold with a 10 nm thick adhesion layer of titanium on a polyimide substrate. The liquid polyimide of PI-5878G (HD MicroSystems$^{TM}$) was spin-coated on a 2 inch silicon wafer to form the substrate. In this work, we fabricated our samples with two different thicknesses, namely 5.5 and 11 $\mu$m. The thickness of the polyimide substrate can be precisely controlled by adjusting the spin rate and curing temperature. AZ5214e image reversal photoresist was patterned using direct laser writing with a Heidelberg DWL 66 laser writer. 200 nm thick Au/Ti was E-beam evaporated followed by rinsing in acetone for several minutes. As a final step, the MM structures patterned on the polyimide substrate were peeled off of the silicon substrate. The as-fabricated 2 inch diameter samples show extreme mechanical flexibility, as shown in figure 1.

Terahertz time-domain spectroscopy (THz-TDS) was used to characterize the metamaterial response. The transmission of the THz electric field, at normal incidence, was measured for the sample and a reference, which in the present case is simply air. The electric field spectral amplitude and phase are calculated through Fourier transformation of the time-domain pulses. Dividing the sample by the reference yields the complex spectral transmission [13]. Prior to measurement, the free-standing MM samples were diced into 1 cm × 1 cm squares and mounted at normal incidence with respect to the THz radiation. All of the measurements were performed at room temperature in a dry (<0.1% humidity) air atmosphere. The THz beam diameter was ∼3 mm, which was significantly smaller than the sample dimensions.

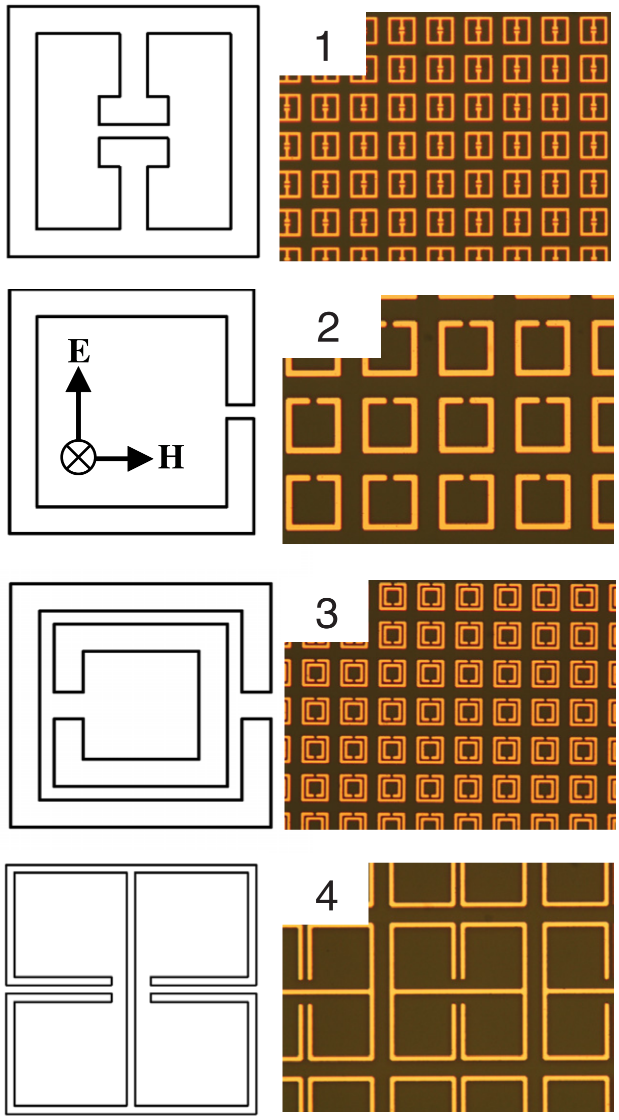


**Figure 2.** Designs and photographs of the polyimide supported metamaterials. The corresponding dimensions are listed in table 1. The arrows labelled $E$ and $H$ show the electric and magnetic field directions with respect to the orientation of all of the SRRs.

THz-TDS measurements were first carried out on a series of substrates without metamaterials to characterize the complex refractive index, $n = n_r + in_i$, of the polyimide samples. Films with thicknesses of 5.5, 11, 60 and 160 $\mu$m were fabricated using spin-coating as described above. For each of these films, a frequency independent refractive index $n_r = 1.8 \pm 0.05$ was experimentally determined (from 0.2–2.5 THz). For all of the samples measured $n_i$ was also frequency independent. However, there was considerable variability in the magnitude of $n_i$ ranging from 0.026 up to 0.150. We attribute this variation to imperfections in the polyimide substrates resulting in scattering which yields an apparent increase in $n_i$. The larger values were obtained from

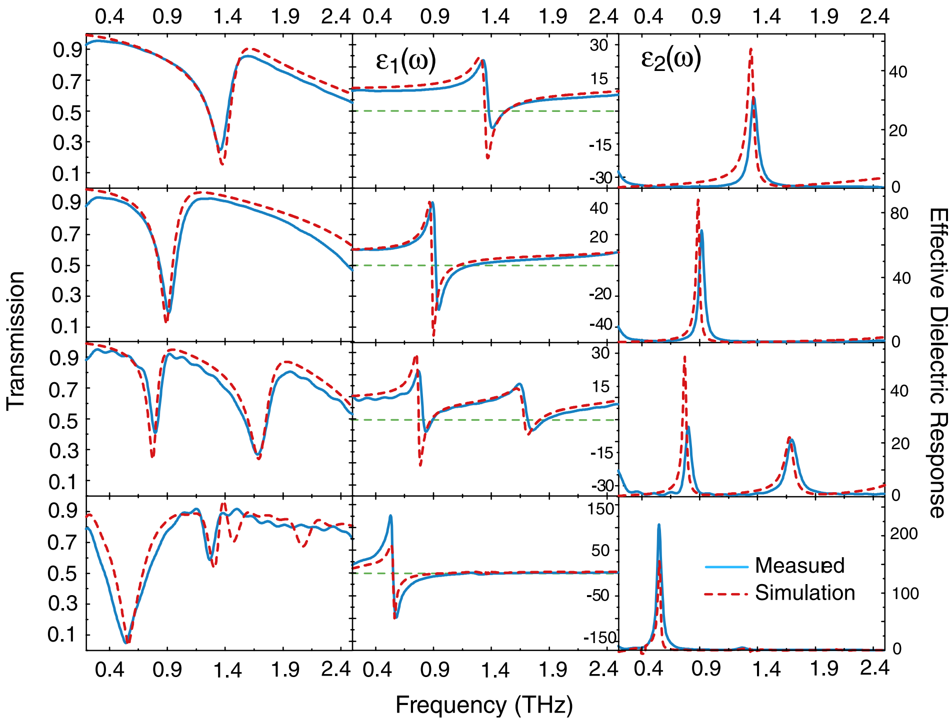


**Figure 3.** The left panels show the experimentally measured transmission for the corresponding samples in figure 2. The solid (blue) line is experiment, and the broken (red) line is the simulated transmission. The middle panels show the real part of the dielectric response and the right panels show the imaginary part. The blue lines are determined from the experimental data and the red lines are determined from the simulations.

the thicker samples consistent with this interpretation. For the thinnest films, the value was typically $n_i = 0.04$ and the field transmission was greater than 0.95 from 0.2 to 2.5 THz. We note that at 1.0 THz $n_i = 0.04$ corresponds to a power absorption coefficient of $\alpha = 2\omega n_i/c = 30\,\mathrm{cm}^{-1}$. In short, polyimide serves as a low index low-loss and highly flexible substrate upon which to fabricate THz metamaterials.

Numerous free-standing polyimide/metamaterial samples were fabricated and characterized using THz-TDS. For brevity, we focus on four of these samples. Schematics of the metamaterial particles are displayed in the left column of figure 2 while the right half of the figure shows optical micrographs of portions of the arrays. The samples include purely electric resonators (1 and 4) and canonical split ring resonators (2 and 3).

We note that all of the samples have a resonant effective permittivity with the electric field oriented as depicted in figure 2. For these normal incidence measurements, there is no coupling to the magnetic response as there is no component of the magnetic field perpendicular to the rings. Further, structures 1 and 4 are designed to be non-magnetic for any orientation of the incident field [25]. While samples 2 and 3 are magnetically resonant, it is not possible to determine the effective permeability as this would require measurements at an angle of incidence of nearly 90° which is not possible in a conventional THz-TDS setup. Thus, in what follows, we present measurements of the transmission and effective permittivity.

The experimental and simulation results are displayed in figure 3. The left-hand side shows (blue lines) the

**Table 1.** Dimensions of each free-standing metamaterial particles (all units in $\mu$m): $a$, lattice period; $b$, outer dimension; $t$, thickness of the polyimide substrate; $w$, line width; $l$, length of the gap; $g$, gap distance; for sample 3, the distance between inner and outer rings is 2 $\mu$m.

| Sample | $a$ | $b$ | $t$ | $w$ | $l$ | $g$ |
|---|---|---|---|---|---|---|
| 1 | 50 | 36 | 5.5 | 4 | 8 | 2 |
| 2 | 50 | 36 | 5.5 | 4 | 4 | 2 |
| 3 | 50 | 36 | 5.5 | 4 | 4 | 4 |
| 4 | 250 | 180 | 11.5 | 6 | 67 | 6 |

experimentally measured field transmission as a function of frequency. The resonances are of quality equal to those previously measured on rigid semiconducting substrates [25]. The red lines are the results of electromagnetic simulations using Microwave Studio where the dimensions listed in table 1 on were used for the SRR elements and the experimentally measured refractive index for polyimide ($n = 1.8 + \mathrm{i}0.04$) was used. The agreement with experiment is quite good.

The middle and right columns of figure 3 show the results of extracting the effective dielectric function ($\epsilon(\omega) = \epsilon_1 + \mathrm{i}\epsilon_2$) for our thin films (the middle column displays $\epsilon_1$ while the rightmost column shows $\epsilon_2$). The determination of $\epsilon(\omega)$ followed the standard approach described in previous publications [14–16] with the exception that the effective dielectric function of the entire MM/polyimide film was determined. Hence, a thickness of 5.5 or 11 $\mu$m, as appropriate, was used. In addition, we determined the dielectric response from the simulated transmission following the approach describe in [26, 27]. The excellent agreement

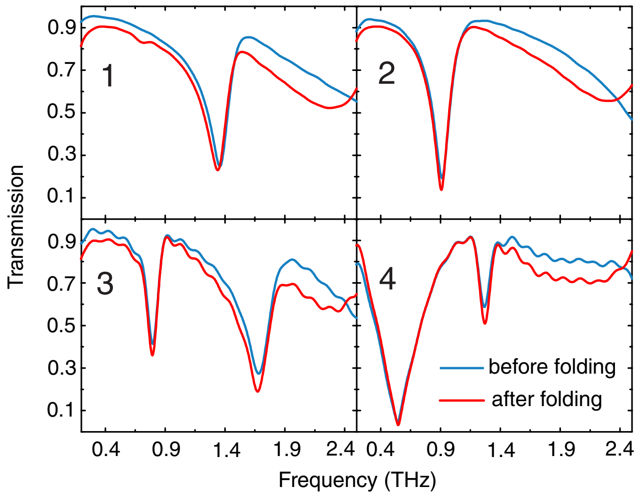


**Figure 4.** The transmission of each of the samples prior to (blue) and after folding and unfolding the samples multiple times (red).

with the experimental data attests to the high quality of the metamaterial samples.

These samples can be wrapped into cylinders with a radius of approximately 3 mm. Harsh environmental tests have been conducted to test the robustness of the free-standing metamaterial samples by rinsing them in organic solutions such as methanol and isopropanol, tearing and tweaking them with considerable mechanical force, and heating them up to 350 °C. No distortion or cracking was observed. In addition, the transmission of the samples was measured in the pristine state and again after considerable folding along multiple directions. Figure 4 shows the results of these measurements indicating virtually no change in the resonant response. These results suggest that such metamaterials can maintain their electromagnetic integrity with fairly vigorous handling.

The use of spin-coating polyimide as a flexible substrate for metamaterials offers numerous possibilities as functional electromagnetic coatings including narrowband filters or absorbers. Importantly, the polyimide thickness can be controlled with sub-micrometre precision and, further, the total substrate thickness can be substantially less than the lateral unit cell dimensions which facilitates the incorporation into multilayer (potentially heterogeneous) structures. The flexible substrate is of sufficiently low loss to enable the construction of multilayer samples including, potentially, perfect absorbers, negative index materials and THz electromagnetic cloaks. As described above, part of the substrate loss arises from scattering due to imperfections. Such losses may be further reduced with improvements in the spin-coating process.

In summary, flexible resonant terahertz metamaterials built on ultrathin highly flexible polyimide substrates have been designed, fabricated and measured. These results pave the way for creating numerous multilayered non-planar electromagnetic composites.

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