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Terahertz time-domain spectroscopy studies of subwavelength hole arrays in metallic films

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Abstract

We studied the phenomenon of ‘anomalous transmission’ in various subwavelength hole arrays in metallic films, using the terahertz (THz) time-domain spectroscopy technique. The perforated samples were heavily doped polypyrrole conducting polymer films patterned with hole arrays in the form of square Bravais lattice. The enhanced transmission in perforated heavily doped polypyrrole films exhibit characteristic properties that can be tuned through chemical synthesis and alteration of the doping level, allowing novel optical devices to be fabricated in the THz spectral range.

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Keywords: Conducting polymers; Terahertz time-domain spectroscopy; Perforated metallic films; Anomalous transmission; Surface waves

1. Introduction

Since the discovery of the ‘anomalous transmission’ phenomenon through a two-dimensional (2D) subwavelength aperture array perforated on optically thick metallic films [1], numerous studies have been performed to understand fundamental properties and explore potential device applications [1–9]. Originally, this ‘anomalous transmission’ was explained as a surface plasmon polariton (SPP) assisted phenomenon [1]; however recently, the original explanation of the phenomenon of ‘anomalous transmission’ was challenged by introducing alternative models [6,7] and underlying mechanism is still actively debated. Here we report the observation of ‘anomalous transmission’ of terahertz (THz) radiation through exotic plasmonic hole arrays such as square Bravais lattice perforated on heavily doped organic conducting polymers films [10]. We succeeded in measuring ‘anomalous transmission’ with profound impact on the physics of doped organic polymers.

In the THz time-domain spectroscopy (THz-TDS) method (see Fig. 1), the properties of a single-cycle electromagnetic transient transmitted through a structure are measured using a pump–probe detection scheme with sub-picosecond temporal resolution. In contrast to conventional optical measurements where the transmitted optical power is measured, THz-TDS allows for the direct measurement of the THz transient electric field that yields both amplitude and phase information [11]. We can also monitor the time evolution of the launched surface waves, which constitutes an advantage for studying plasmonic lattices [9].

Doped conducting polymers such as polyacetylene, polyaniline and polypyrrole, show a metal–insulator transition at high doping levels of a few percent [12]. Among the class of conducting polymers, polypyrrole [PPy] heavily doped with PF_6 [PPy(PF6)] exhibits one of the highest conductivities, while maintaining very good stability in air [13,14]. This high conductivity (>100 S/cm) is maintained even at low temperatures. It has been postulated that heavily doped PPy(PF6) has two plasma frequencies, where the lower frequency seems to be caused by a Drude free electron dielectric response with a plasma

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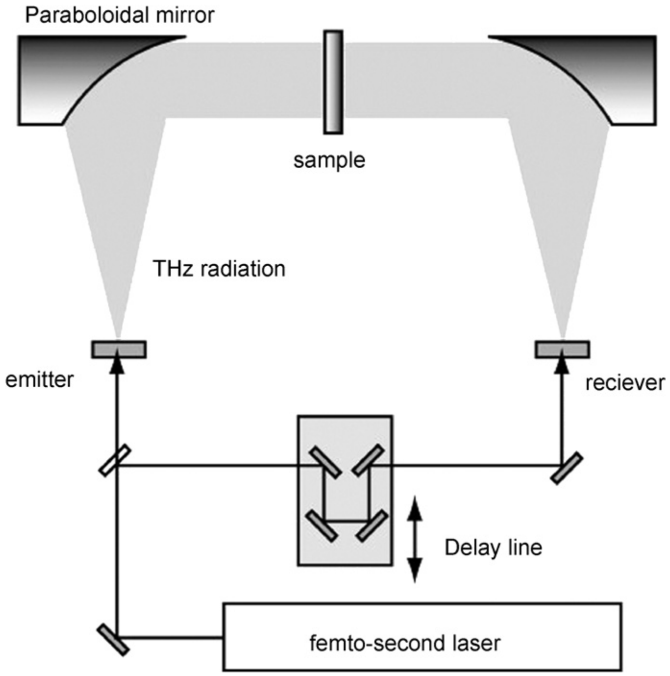

Fig. 1. Schematic representation of the THz-TDS optical setup.

frequency in the THz spectral range. Our finding of anomalous transmission in perforated doped polymer films shows that SPP excitations can be formed on the surfaces of metallic conducting polymers; thus in this sense these novel organic metallic films behave as regular metals.

2. Experimental

2.1. Sample preparation

PPy films were polymerized and doped electrochemically with PF_6 molecules at -40 °C. The molecular structure of PPy is shown in the inset of Fig. 2. The room-temperature DC conductivity of the heavily doped PPy(PF6) was measured to be ~200 S/cm. For the 2D aperture array fabrication the PPy(PF6) films were peeled off the electrodes following polymerization in the form of free-standing films with thicknesses of ~25 μm. The 2D hole arrays were then patterned on the polymer films using a pulsed excimer laser machining system (Optec, Micro-Master) in a geometry of a square Bravais lattice. We fabricated two samples on the PPy films with different periodicities of 1 and 1.5 mm, with a hole diameter of 0.5 and 0.75 mm, respectively. The fractional aperture area was thus kept constant at ~20%.

2.2. Terahertz time-domain spectroscopy (THz-TDS) measurements

We used a conventional THz-TDS set-up (Fig. 1) to characterize the complex dielectric constants of PPy(PF6) films and transmission through the perforated metallic films in the spectral range between 50 GHz and 0.6 THz. [15]. In our setup photoconductive devices were used for both THz generation and coherent detection. The samples were placed at the center of two off-axis parabolic mirrors used to collect, collimate, and focus the THz electromagnetic (EM) radiation. The linearly polarized THz EM beam was normally incident on the film surface. For normalization purposes reference spectra were measured before placing the samples in the THz beam path.

A unique feature of the THz-TDS technique is that it allows for a direct measurement of the transient electric field, so that both amplitude and phase information are obtained simultaneously. We Fourier-transformed the transient photocurrent response to the frequency domain, and evaluated both real (n_r) and imaginary (κ) refractive indices spectra without the need for Kramers–Kronig analysis. Using these spectra analyses, we also obtained the spectra of the transmission, reflectivity, as well as the real and imaginary dielectric constants from known relations between the various optical constants and the complex refractive index $n = n_r + i\kappa$.

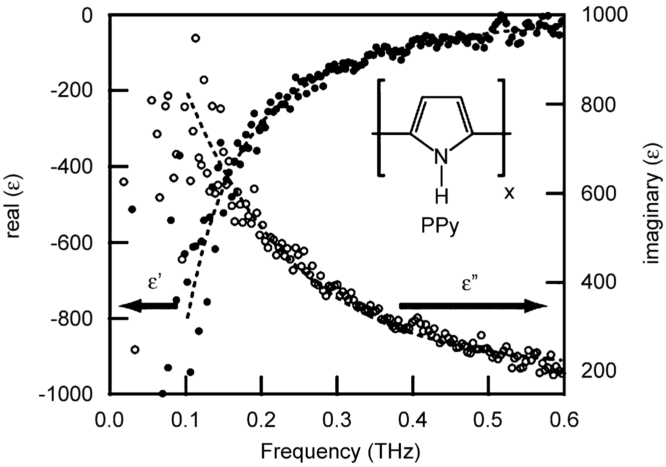

Fig. 2. THz spectra of the real (ε'; filled circles) and imaginary (ε''; empty circles) components of the dielectric constant for the heavily doped PPy(PF6) free standing film 25 μm thick. The lines through the data points are guide to the eye. Inset: molecular structure of the PPy polymer.

3. Results and discussion

We first measured the complex dielectric constant spectrum of the unperforated PPy(PF6) film. Fig. 2 shows the measured real (ε') and imaginary (ε'') dielectric constant spectra of the heavily doped PPy(PF6) film in the THz spectral range. We indeed found that ε' is negative over the THz spectral range of interest, and this corresponds to 'metallic' behavior. Therefore the PPy(PF6) film should support SPP excitations in the measured spectral range.

Next, we examined the transmission characteristics of 2D periodic subwavelength aperture arrays fabricated on the PPy(PF6) films. Fig. 3 shows a typical transient response of THz radiation $E(t)$ through the perforated