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Nonlinear Plasmonic Metasurface Terahertz Emitters for Compact Terahertz Spectroscopy Systems

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achievable with conventional THz emitters. However, they were driven to date only by amplified laser systems, which are expensive and have a large footprint, thus limiting the range of their potential applications. Here we study for the first time the possibility to drive metasurface emitters by low-energy near-infrared femtosecond pulses. We observe broadband THz emission from 40 nm thick metasurfaces and achieve near-infrared to THz conversion



efficiencies as high as those of 2500-fold thicker ZnTe crystals. We characterize the THz emission properties and use the metasurface emitter to perform a spectroscopic measurement of α -lactose monohydrate. These results show that nonlinear plasmonic metasurfaces are suitable for integration as emitters in existing compact THz spectroscopy and imaging systems, enhancing their functionalities, and opening the door for a variety of new applications.

KEYWORDS: terahertz (THz), nonlinear metasurface (NLMS), plasmonic metasurface antenna (PMA), low-energy excitation, spectroscopy

In the past few decades, terahertz (THz) electromagnetic waves, in the frequency range of 10¹¹–10¹³ Hz, have found a wide variety of interesting emerging scientific and technological applications. They have been used, for example, to probe the temporal dynamics of high temperature superconductors,¹ for strong coupling of cavity modes with collective intermolecular vibrations,² for controlling molecular orientation,³ coherent manipulation of semiconductor quantum bits,⁴ and also for probing and manipulating antiferromagnetic spin waves.⁵ In addition, since many molecular systems show unique spectral THz signatures, from rotational levels of small molecules, through intramolecular vibrations of macromolecules and intermolecular vibrations of hydrogen bonded molecular crystals, THz spectroscopy is a promising tool for biomedical⁶ and security⁷ applications.

In spite of all these important emerging applications, the generation, detection, and control of THz waves have proved to be highly challenging.⁸ Electronic circuits, while useful in the microwave frequency regime, suffer from losses and parasitic effects that greatly reduce efficiency when operating at hundreds of gigahertz.⁹ On the other hand, conventional techniques for generation, detection, and manipulation of light in the infrared and visible regime, are not applicable for the low energy of THz photons.¹⁰

The pursuit of good THz emitters has led to the development of numerous innovative solutions. These include THz quantum cascade lasers,¹¹ photoconductive antennas

(PCAs),¹² synchrotron radiation,¹³ optical rectification (OR) in inorganic or organic nonlinear crystals,^{14,15} and three wave mixing in air,¹⁶ to name a few. However, the existing sources allow limited control over the properties of emitted THz radiation, for example, the beam shape and temporal profile. This requires the use of additional THz components that, in addition to being scarcely available, add further losses and aberrations. Therefore, there is still a need to develop broadband THz emitters that require a low energy driving source, work at room temperature, allow to obtain control over the characteristics of the emitted THz radiation, and can be integrated in compact systems. One promising type of THz emitters that has attracted attention lately, is plasmonic metasurface THz antennas (PMAs).

Starting with the work of Kadlec et al.¹⁷ it has been shown that broadband THz pulses are emitted when illuminating metallic surfaces with near-infrared (NIR) femtosecond lasers, where symmetry breaking at the surface plays a crucial role in the conversion process.^{18–22} Significant enhancement of the

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conversion efficiency was observed when the surfaces were structured at the nanoscale.¹⁹ This enhancement has been attributed to the coupling between the optical fields and localized plasmonic modes in the metal.^{20,21}

Lately, metasurfaces consisting of gold split-ring resonators (SRRs) were shown to be relatively efficient broadband THz emitters, generating THz pulses at the same order of magnitude as conventional, much thicker, ZnTe crystals.²³ Moreover, the ability to shape the spatiotemporal behavior of emitted THz pulses using nonlinear metasurfaces was demonstrated and studied,²⁴⁻²⁷ opening the door to functionalities unavailable by other means of THz generation. Yet, all the demonstrations of THz generation by metasurfaces to date used amplified laser systems. Such lasers are expensive, heavy, and have a very large footprint, consequently drastically limiting the potential THz applications. In addition, amplified systems usually operate at relatively low repetition rates of several kHz, which may put additional constraints on the measurement speed. Compact laser oscillators, on the other hand, are economical, portable, and widely accessible. However, previous works that used laser oscillators to excite metallic nanostructures, noted conversion efficiencies much lower than those of conventional generation methods.^{19,28} Here we study the generation of THz pulses using an SRRbased PMA excited with a laser oscillator emitting nanojoule femtosecond pulses. We measure output signals as high as those generated by OR in a 0.1 mm thick ZnTe crystal and characterize their polarization and dependence on pump power. We then use the PMA as an emitter for performing spectroscopic THz measurements of α -lactose monohydrate powder.

The PMA that we used consists of a uniform array of 40 nm thick gold SRRs, fabricated using conventional electron beam lithography (see Methods). An illustration of a typical SRR's dimensions and a scanning electron microscope (SEM) image of the sample are presented in Figure 1a and b, respectively.



Figure 1. (a) Typical SRR dimensions. (b) SEM image of a small area of the PMA showing the SRRs. (c) Spectral transmission through the PMA when excited with light polarized along the SRR base (*x*-axis), showing a dip at ~1530 nm.

Figure 1c presents the linear spectral transmission through the metasurface, indicating a plasmonic resonance centered at \sim 1530 nm for excitation with light polarized linearly along the SRRs base (defined as the *x*-axis).

THz generation and detection were performed using a standard time-domain spectroscopy (TDS) setup, as depicted in Figure 2. To drive the THz generation, we used the output

of an optical parametric oscillator pumped by a Ti:sapphire femtosecond laser (Chameleon Compact OPO). The system emits pulses (\sim 170 fs) with energies of several nanojoule per pulse at 1500 nm, which fits well with characteristics of compact and affordable femtosecond fiber lasers. The pump was weakly focused on the THz emitter (PMA or ZnTe) with a 400 mm lens. See Methods for a more detailed description.

The orange line in Figure 3a shows a typical THz pulse emitted from the PMA using laser pulses with an energy density of 2.8 μ J/cm² per pulse. It can be seen that single cycle pulses are emitted with a duration of \sim 1 ps. We compared the emission from the PMA to that from a 0.1 mm thick ZnTe emitter; while ZnTe is optimally phase matched for excitation wavelengths of 800 nm, at 1500 nm and for THz frequencies relevant to this work, the coherence length of ZnTe (~0.2 mm or longer) is larger than the thickness of the crystal that was used. A typical THz field pulse emitted from the ZnTe crystal, under the same pumping conditions as the PMA, appears as the blue line in Figure 3a. The THz signal in this case showed an initial single cycle THz waveform with an oscillating tail. Notably, it can be seen that the 40 nm thick PMA emits a THz field nearly equal in amplitude to that of the 0.1 mm thick ZnTe crystal. In addition, it is important to note that the THz field emitted from the PMA showed no degradation with continuous pumping of over 3 hours. The orange and blue lines in Figure 3b show the THz spectra emitted from the PMA and the ZnTe crystal, respectively, as were obtained by Fourier transforming the time domain signals, showing a bandwidth of ~1.5 THz with a noise level lower than -30 dB. The spectral dip in the ZnTe spectrum corresponds with the oscillating tail seen in Figure 3a and may be explained by a Fabry-Pérot mode of the crystal.

To put these results into context, it is worth noting that when working with laser oscillators, PCA emitters are usually the tool of choice,²⁹ showing conversion efficiencies much larger than OR in nonlinear crystals.³⁰ Also, novel sources like spintronic devices are recently explored as highly efficient and broadband sources.^{30,31} In this sense, although the conversion efficiency we have measured from the PMA is comparable to that of a ZnTe emitter; at this point it is still considerably lower than the efficiency achieved with active sources in oscillator systems. However, the unprecedented control PMAs allow over the beam shape, as well as metasurfaces' tunability to different pump frequencies, may make them attractive for use as THz sources in low energy setups, even at the expense of THz power. Moreover, theoretical works suggest that NIR to THz conversion efficiencies of PMAs may be improved by a few orders of magnitude using collective effects,³² potentially allowing for future PMA THz sources that compare to or even surpass state of the art PCAs in efficiency.

To characterize the THz emission from the PMA, we measured the dependence of the THz field on the pump power. Using a half-waveplate and a polarizing beam splitter, the pump power was varied. This affected only the amplitude of the emitted THz pulse, without modifying its shape. The dependence of the THz peak to peak amplitude on the pump power is depicted in Figure 4a. A linear dependence is apparent, indicating a structural second order nonlinear process. In addition, we examined the polarization of the emitted THz field by placing a wire-grid polarizer between the PMA and the first parabolic mirror. The measurements presented in Figure 4b show that the emitted THz field is linearly polarized along the *y*-axis, perpendicular to the SRR

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Figure 2. TDS experimental setup. The illumination source was an OPO emitting NIR pulses (\sim 170 fs, 1500 nm) at a repetition rate of 80 MHz, fed by a Ti:sapphire laser emitting 808 nm pulses. The THz emission was collected by a fiber coupled PCA that is triggered by a portion of the NIR pulse. The Teflon lens collimated the THz light and prevented the pump pulse from reaching the PCA. Temporal scanning by the delay stage allowed to measure the time domain shape of the electric field of the THz pulse.



Figure 3. (a) THz signal emitted from the PMA (orange) excited along the SRRs' base (measured perpendicular to pump polarization) and from a 0.1 mm thick, (110) cut ZnTe crystal (blue), excited along its $[\overline{111}]$ axis (measured parallel to the pump's polarization). Both time domain signals are normalized by the peak amplitude of the THz pulse emitted from the ZnTe crystal. (b) Emission spectra of the signals shown in (a), obtained by Fourier transform and normalized by the maximum spectral power of the ZnTe crystal.



Figure 4. (a) Dependence of the amplitude of THz light generated by the PMA on the pump power (black circles) and a linear fit (orange line). A linear dependence is apparent, indicating a second-order process. (b) Polarization of the THz light generated by the PMA (black circles) and a fit to a $\cos^2(\theta_P)$ dependence (orange line). θ_P is the angle of the polarizer's transmission axis measured from the *y*-axis (perpendicular to the SRR base).

base. The linear polarization is confirmed by fitting the emission profile to a $\cos^2(\theta_{\rm P})$ function, where $\theta_{\rm P}$ is the polarizer transmission axis angle relative to the *y* axis. The projection of the \hat{y} -polarized THz field, first on the polarizer transmission axis, and then on the PCA measurement axis (\hat{y}) , leads to the squared cosine dependence.

Finally, to explore the suitability of the PMA for THz spectroscopy applications, we performed a spectroscopic measurement of a \sim 0.7 mm thick powder of α -lactose

monohydrate (47287-U, Sigma-Aldrich), using the PMA as the THz source. The α -lactose powder was inserted into a nylon bag, and the sample was placed after the collimating Teflon lens (see Figure 5a). Figure 5b shows the time domain

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Figure 5. (a) Arrangement of the TDS setup for a spectroscopic measurement of α -lactose powder (full setup is displayed in Figure 2). (b) Time domain signals of the empty nylon bag (black line) and of the bag with α -lactose powder (green line). (c) (Circles) Absorbance spectrum of a ~0.7 mm thick sample of α -lactose powder, measured with the PMA acting as the THz source. (Solid line) Lorentzian fit to the absorbance, showing the known absorption line due to collective intermolecular vibrational transitions at 0.53 THz.

signals of two transmission measurements, one of the empty nylon bag and one of the bag with α -lactose powder. The absorbance of the powder was calculated from the two spectra, Figure 5c shows the extracted absorption line at 0.53 THz. This is the lowest lying THz absorption line of α -lactose due to collective intermolecular vibrational transitions.³³ The two additional absorption lines at 1.2 and 1.4 THz could not be discerned, neither with the PMA nor with the ZnTe emitter. Measuring the two additional lines may either require systems with higher signal-to-noise ratio, or use of stricter sample preparation protocols,² as the spectroscopic response of granulated solids was shown to depend strongly on sample preparation.³⁴

To conclude, we have studied the possibility to drive THz PMAs by nanojoule femtosecond lasers, which are commonly used in compact THz spectroscopy and imaging systems. The 40 nm thick PMA generated linearly polarized single-cycle THz pulses with a bandwidth of ~1.5 THz and noise level of under -30 dB, with NIR to THz conversion efficiency as high as that of a 0.1 mm thick ZnTe crystal. This allowed us to

perform a spectroscopic measurement of α -lactose powder and identify the familiar absorption line at 0.53 THz. We believe that these demonstrations show that PMAs can be used as sources in compact and affordable THz systems. While various works studied the THz emission process from nanostructured metallic surfaces, the role of different physical mechanisms that lead to THz emission is not yet fully understood.^{20,21,35} Better understanding of the underlying physical mechanisms and how they can be harnessed, together with the optimization of PMA design, for example, by utilizing collective effects on the metasurface,^{32,36} or integrating with additional materials^{37,38} may allow for improvement of conversion efficiencies from such emitters beyond currently reported values. This, along with expansion of the exceptional functionalities available with PMAs,²⁴⁻²⁶ would pave the way for extremely compact THz setups with moderate field amplitudes that allow tailoring the THz pulse to the application needs.

METHODS

Sample Preparation. The PMA was fabricated on glass coated with ~20 nm of indium-tin-oxide (ITO). Before fabrication, the substrate was cleaned by sonication in acetone and isopropyl alcohol and dried (throughout the fabrication process, drying was done using N₂ gas). After cleaning, the substrate was spin-coated with polymethyl methacrylate (PMMA A4) and baked on a hot plate for 1 min at a temperature of 180 °C. The SRR array was written in the resist using electron-beam lithography (Raith 150-II, 20 kV), with the ITO coating acting as the conducting layer required for the lithographic process. After that, the sample was developed by immersion in a solution of MIBK/IPA (1:3) for 1 min at a temperature of 4 °C and dried. A 3 nm Ti adhesion layer was evaporated on the developed sample, followed by evaporation of a 37 nm layer of Au. Lift-off of the resist was performed by immersion in acetone, finally drying the prepared sample.

Experimental Setup. For the generation and measurement of THz pulses we used an optical parametric oscillator (Chameleon OPO VIS) emitting ~170 fs pulses centered at a wavelength of 1500 nm, with a repetition rate of 80 MHz. The IR pulse was split into two paths, using a half-wave plate and a polarizing beam splitter, with ~280 mW of the light passing to the pump path and ~ 30 mW to the probe path. The pump passed through a delay stage and an optical chopper modulated at 1.9 kHz. After the chopper, a half-waveplate allowed setting the polarization axis of the pump, and the beam was focused with a 400 mm lens onto the sample leading to pulse energy densities of up to 2.8 μ J/cm². The THz pulse that was emitted from the samples passed through two gold coated off-axis parabolic mirrors with a diameter of 50.8 mm and focal length of 50.8 mm, arranged in a 4f configuration. At the output of the 4f system, the THz signal was collimated with a 25.4 mm hemispherical Teflon lens, which also blocked the NIR pulse, and then focused into a PCA (Menlo systems TERA15-RX-FC). Meanwhile the probe pulse was coupled to a 1 m long fused silica single mode fiber to excite carriers in the PCA that was set to measure the component of the THz field polarized along the y-axis.

For measurements of the THz signal emitted from the PMA (or ZnTe) the pump was linearly polarized along the SRR base (or $[\overline{1}11]$ axis of the crystal) and the PCA orientation allowed measuring the THz field component perpendicular (or parallel) to the pump. In addition, the PMA was always positioned so that the pump light first passed through the glass

substrate, then through the ITO coating and finally reached the metasurface. Measurements with the PMA placed in reverse yielded an order of magnitude weaker THz signals. In measurements comparing between the THz field emitted from the PMA and a ZnTe crystal, the system was first aligned to optimize the THz signal measured from the ZnTe crystal. Then the crystal was replaced with the PMA without any modification to the alignment except for rotation of the pump polarization using the half-waveplate.

Measurements of the current in the PCA were done using a lock-in amplifier (Stanford Research Systems, SR830) locked to the chopper modulation frequency. THz generation and detection were done in ambient air, without purging the setup. In the energy densities used in the experiment no degradation of THz emission was observed. However, when using a 250 mm lens instead of 400 mm (energy densities of 7.3 μ J/cm²), the signal from the PMA degraded quickly in the first few minutes of continuous pumping due to laser damage to the sample.

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Notes

The authors declare no competing financial interest.

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