Germanium Plasmonic Nanoantennas for Third-Harmonic Generation in the Mid Infrared

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***Abstract*—We explore the nonlinear optical properties of plasmonic semiconductor antennas resonant in the mid infrared. The nanostructures are fabricated on silicon substrates from heavily doped germanium films with a plasma frequency of 30 THz, equivalent to a wavelength of 10 µm. Illumination with ultrashort pulses at 10.8 µm produces coherent emission at 3.6 µm via third-harmonic generation.**

I. Introduction

Recent advances in semiconductor film deposition allow the growth of heavily doped germanium with effective plasma frequencies of up to 30 THz. This technology paves the way for mid-infrared (MIR) nanoplasmonics with application to integrated telecommunications systems and to precise sensing in the spectral region defined as the vibrational fingerprint of molecules. Characteristics like CMOS compatibility, low electron effective mass and tunable dielectric function give advantage to Ge over metal plasmonics or other semiconductors. [1,2]

In this work, we demonstrate that plasmonic Ge antenna structures are suitable for driving nonlinear optical processes such as third-harmonic generation (THG) in the mid infrared [3] owing to the strong near-field enhancement. These devices act as light emitters constrained at sub-wavelength dimensions and consequently are of high interest for experiments targeting single molecules or other isolated quantum systems [4].

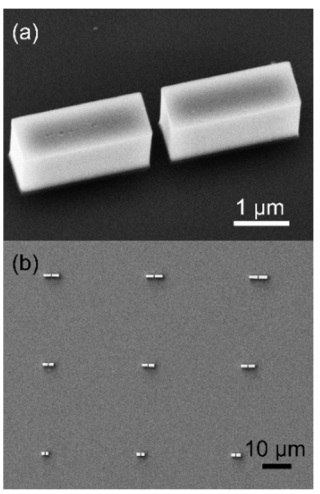

Fig. 1. Scanning electron microscope images of (a) an isolated germanium double rod antenna with an arm length of 2 µm and a gap width of 300 nm on silicon substrate and of (b) an array of 9 antennas with different arm length (top view).

II. Experiment and Results

Doped single-crystalline germanium films with a thickness of 1 µm are grown epitaxially via low-energy plasma enhanced chemical vapor deposition (LEPECVD) on intrinsic silicon substrates. Double rod antenna structures are then fabricated via electron beam lithography (see Fig. 1) to be resonant at the mid-infrared excitation wavelength. Simulations considering the antenna geometry and the material dielectric function close to the plasma edge determine the exact resonance frequency. [5]

The optical characterization system is driven by a Yb:KGW femtosecond laser with a repetition rate of 50 kHz and a pulse energy of 2 mJ. It is equipped with an optical parametric amplifier (OPA) delivering broadband pulses in the near infrared [6]. Subsequently, difference frequency generation between the OPA output and residual fundamental radiation in a GaSe nonlinear crystal produces intense few-cycle pulses tunable in the mid-infrared spectral range from 8 µm to 22 µm wavelength. Depending on the GaSe thickness it is possible to generate either ultrabroadband pulses or narrowband but intense MIR transients (see Fig. 2). The temporal pulse characterization is performed via time-domain electro-optic sampling (EOS). Excitation fields of up to 20 MV/cm are reached in the focus of a dispersion-free Cassegrain-Schwarzschild reflecting objective with a numerical aperture of 0.5. A second, identical objective images the antenna in linear transmission geometry. Selected crystalline dielectric and semiconductor filters as well as a grating monochromator are employed for dichroic spectral filtering.

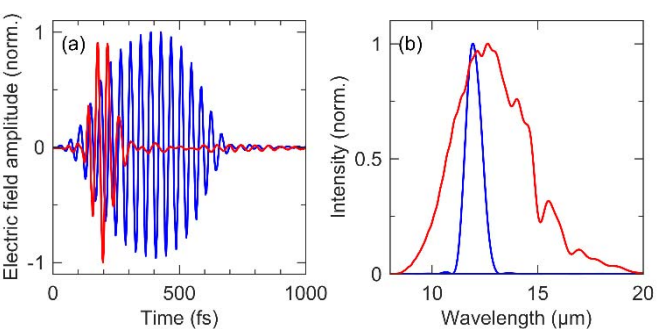

Fig. 2. (a) EOS time trace of two MIR pulses, centered approximately at a wavelength of 12 µm, obtained by difference frequency generation in 140-µm-thick GaSe (red line) and 1.22-mm-thick GaSe (blue line); (b) the respective intensity spectra calculated via Fourier transform.

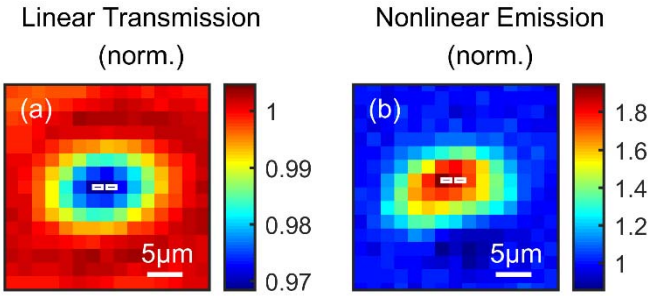

Fig. 3. (a) Linear transmission map of a 2.75-µm-long double rod antenna illuminated with a central wavelength of 10.8 µm including a sketch of the structure (white); (b) spatially resolved third harmonic generation normalized to the silicon substrate background emission.

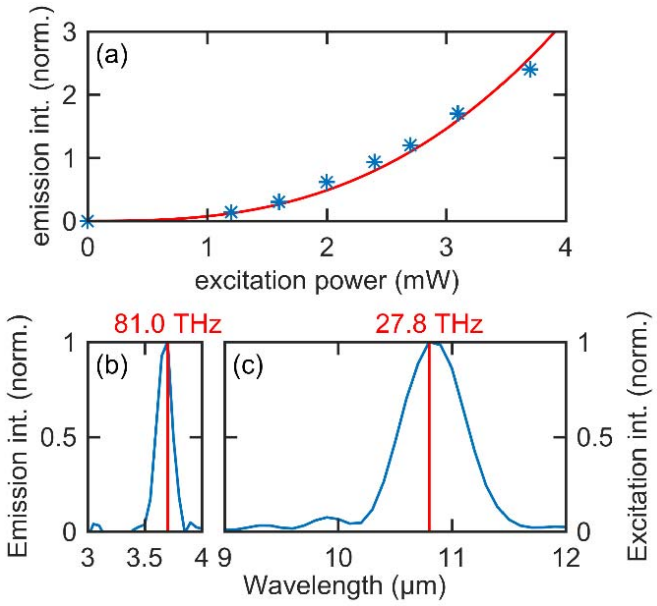

Fig. 4. (a) Power dependence of nonlinear emission of a single Ge antenna (blue: measurement, red: fitting curve ~ $x^{2.72}$); (b) and (c) normalized THG and excitation intensity spectra, respectively.

A liquid nitrogen cooled mercury cadmium telluride detector collects the emission while the sample is scanned through the common focus. This allows addressing single antennas and mapping their linear and nonlinear cross-sections. We study different antenna geometries, sizes and material doping levels.

Fig. 3(a) demonstrates the transmission image at the fundamental wavelength with the increased scattering of an isolated nanostructure while Fig. 3(b) plots the corresponding THG emission that is strongly enhanced at the antenna position with respect to the substrate. The experimental variation of the excitation intensity reveals the power dependence (Fig. 4(a)) typical for a third-order optical nonlinearity. Fig. 4(b-c) show the spectral characterization of the third harmonic photons that are generated at 3.6 µm in comparison to the fundamental excitation set at 10.8 µm.

III. Summary

In this study we show third-harmonic generation from isolated, semiconductor plasmonic structures under intense excitation with femtosecond pulses. Highly doped, crystalline germanium is the ideal material for the mid-infrared spectral range. Power dependence, geometric resonance conditions and emission spectra are fully characterized. Thanks to the large near field enhancement, such nonlinear optical antennas generate ultrashort light sources constrained below the diffraction limit. This work opens interesting perspectives for experiments targeting molecular resonances or other isolated quantum systems in the MIR frequency range.

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