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Near Field Enhancement for THz Switching and THz Nonlinear Spectroscopy Applications

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Abstract: Slits in thin metal sheets and split-ring resonators (SRR) featuring gaps on the micro- or nano-scale are shown to be a promising tool for THz switching or THz nonlinear spectroscopy applications. Both structures show strong field enhancement in the gap region due to light-induced current flows and capacitive charging across the gap. Whereas nano-slits allow for broadband enhancement the resonant behavior of the SRRs leads to narrowband amplification and results in field enhancement of tens of thousands. This property is particularly beneficial for the realization of nonlinear THz experiments.

Keywords: THz nonlinear spectroscopy

Introduction

The last decades have seen significant progress in the development of terahertz technologies in a wide variety of research fields and applications such as chemical recognition, material inspection, or security control. One of the major and yet unmet limitations is that, compared to the optical regime, the pulse energies supplied by current THz sources are still rather limited preventing the advent of THz nonlinear spectroscopy. The highest average THz power levels currently available come from large-scale electron accelerators.[1] Using table-top sources generation of high energy THz pulses has been demonstrated using for example large area photo-conductive switches,[2] frequency mixing in laser-generated plasmas,[3] or optical rectification in nonlinear crystals.[4]

In order to extend THz experiments into the nonlinear regime, *i.e.* for electric or magnetic switching applications or nonlinear spectroscopy, the quantity to be optimized is essentially the electric field strength E since a nonlinear process of order n scales with E_n. The field strength is linked to the pulse energy Q through the relation $E = \sqrt{(Q/(\Delta\tau\ A))}$ where $\Delta\tau$ is the pulse duration and A denotes the cross-sectional beam area. The available pulse energy is naturally limited by the THz system at hand and the pulses are typically already single-cycle so that one cannot increase the field strength by adjusting the parameters Q and $\Delta\tau$. The cross-sectional beam area can be minimized through tight focusing typically with parabolic mirrors. However, the diffraction limit imposes a lower boundary on A thereby restricting the obtainable field strength. Fig. 1a) and b) show the peak electric field strength and peak magnetic field that is obtained when a single-cycle pulse is focused to a diffraction limited focal spot by a F#=1 optics.

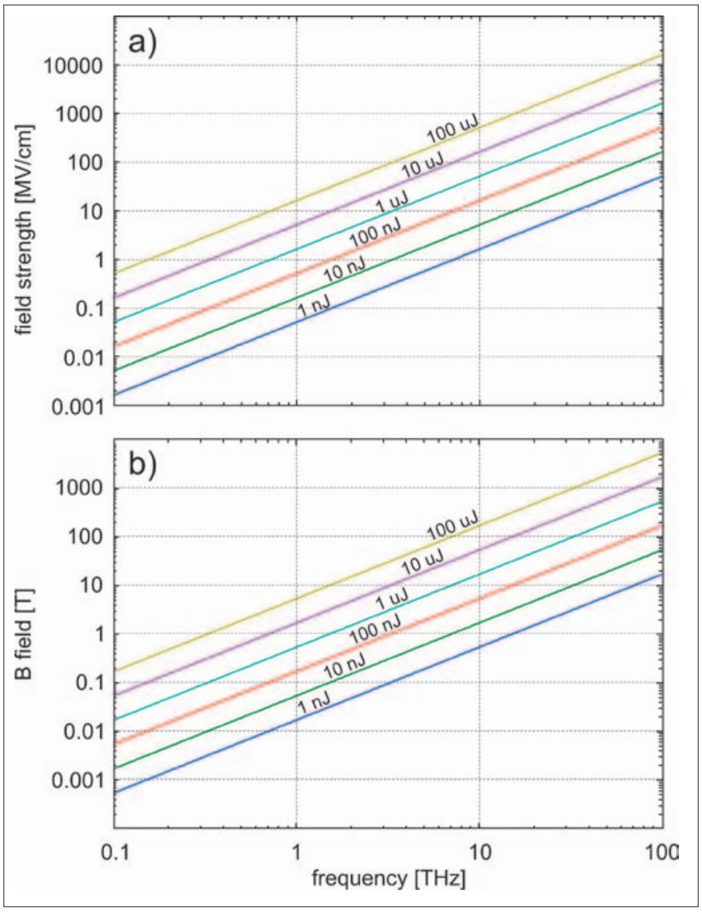

Fig. 1. a) Electric field strength and b) magnetic field as a function of frequency for various energies of single-cycle pulses focused at the diffraction limit.

Both the electric field strength and the magnetic field scale with frequency; it is much easier to reach high fields for high frequencies.

Recently, several reports have shown that the limitations due to diffraction limited focusing can be overcome by using

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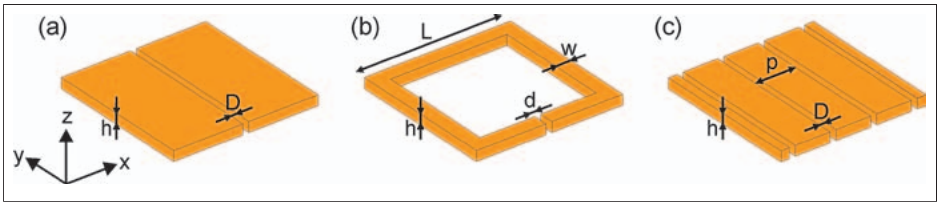

Fig. 2. a) Single nanoslit, b) split ring resonator, and c) 1D array of nanoslits.

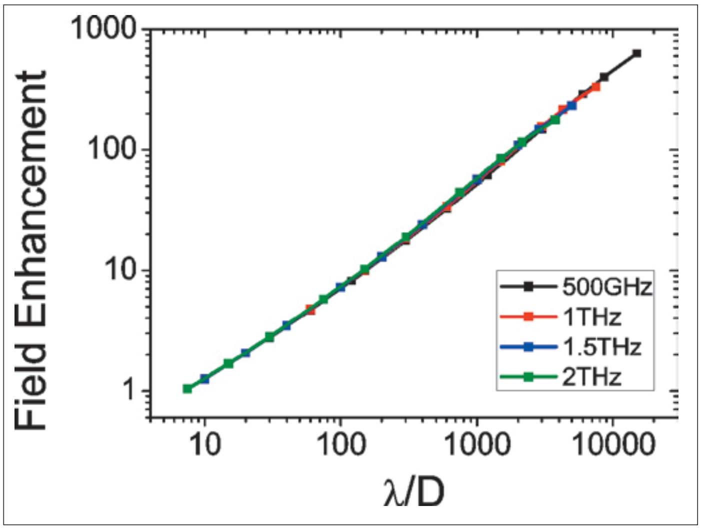

Fig. 3. Field enhancement in a single nanoslit of width *D* versus λ/D for four different frequencies.

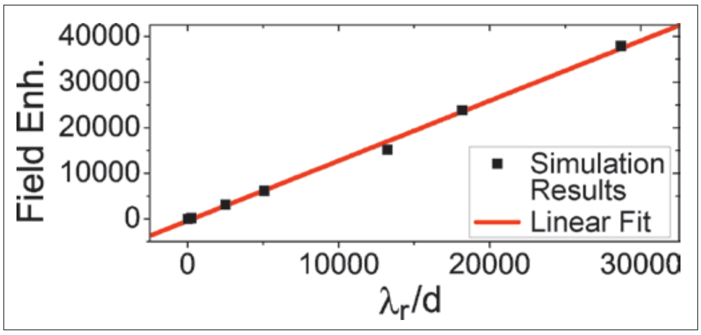

Fig. 4. Field enhancement in a single SRR versus λ_r/D for a number of frequencies. The solid line is a linear fit to the simulated data points.

metallic nano-structures that collect the incident radiation and focus it in a sub-wavelength volume leading to strong field enhancement. This concept was introduced by Seo and co-workers[5] who have reported that a single nano-slit in a thin gold film (Fig. 2(a)) may act effectively as a nano-capacitor. The incident radiation induces a current flow on the metal surface that leads to an accumulation of charge carriers in the gap region. This capacitive charging in turn results in an in-gap enhancement of the electric and magnetic field by orders of magnitude. For a 70 nm wide slit and a frequency of 0.1 THz, a field enhancement factor on the order of 1000 was reported by the authors.

In a recent report we proposed a different geometry in order to further increase the obtainable field enhancement. The structure consists of SRRs[6] that are resonant in the THz regime and that, compared to their side length, feature extremely small gaps (Fig. 2(b)). At the fundamental resonance strong circulating currents are excited in the ring that lead to a build up of charge across the gap.[7] Due to the resonant behavior the resulting field strengths in the gap region are significantly higher than those for the non-resonant nano-slits. For SRRs with gap widths on the nanoscale giant enhancement factors of tens of thousands are obtained.

Enhancing the field strength is one side of the medal; the other is to maximize the integrated nonlinear response. To that end the overall size of the volume comprising the high field strengths has to be maximized. We therefore investigate how the nonlinear response can be maximized by adjusting the structural parameters of both nano-slits and SRRs.

Numerical Simulations

The numerical simulations we show are based on the finite element method (FEM)[8] using a commercial software package.[9] They were carried out in both the frequency- and the time domain. While the frequency-dependent simulations present information on the obtainable field enhancement factor, the time-dependent simulations are employed to evaluate the nonlinear material responses. Modeling of the slit structures could be carried out in two dimensions assuming infinite slit lengths whereas for the SRRs a three-dimensional geometry was employed. The slit structures were positioned in the center of a rectangular (box shaped) simulation domain and the refractive index of the surrounding dielectric was set to unity. We chose to investigate the interaction of the THz waves and pulses with slit and SRR arrays rather than single structures. For this purpose the corresponding boundaries were set to periodic conditions and the size of the simulation domain was chosen according to the periodicity.

Single Nano-Slit

For the sake of completeness we briefly summarize the results published in the literature for a single nano-slit as schematically shown in Fig. 2(a). The slit is illuminated with a normally incident electromagnetic field with the electric field vector oriented across the gap. The field enhancement factor is determined from frequency dependent simulations through the ratio of fields with and without the slit being present. The simulations show that for small slit widths D, *i.e.* λ>D, the field enhancement factor scales roughly linear with λ/D independent of frequency as shown in Fig. 3.

The curves corresponding to different frequencies overlap, which shows that the funneling is a purely geometric effect and that the smaller the slit on the scale of the wavelength the larger is the field enhancement factor. For the smallest considered slit width (D = 40 nm) and frequency (ν = 100 GHz) a maximum field enhancement factor around 3000 is found.

Single Split Ring Resonator

SRRs (see Fig. 2(b)) show an even more pronounced resonant response than nano-slits. Furthermore the spectral position of the SRR resonances can be controlled by adjusting the side length L.[10,11] The fundamental resonance can be excited through either an electric field component parallel to the gap, a magnetic field component perpendicular to the SRR plane, or by a combination of the two.[12,13] Here, we consider E-field excitation where the E, H, and k triad of the incoming field is oriented along the x, y, and z axes. We use L = 300 µm to obtain resonances at the low frequency

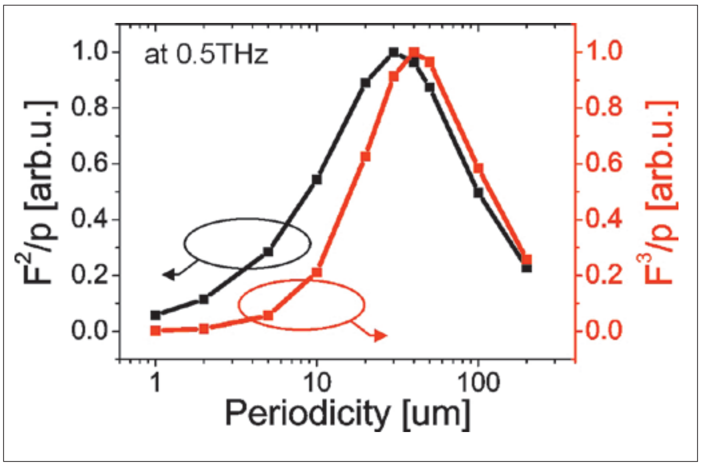

Fig. 5. Product of the volume of high field strength and the field enhancement to the 2nd and 3rd power versus periodicity (black and red symbols, respectively).

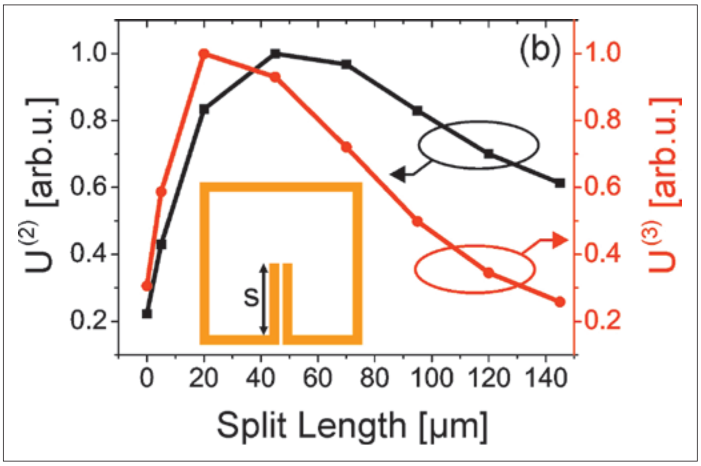

Fig. 6. Field enhancement to the 2nd and 3rd power integrated over the volume of high field strength versus split length (black and red symbols, respectively).

side of the THz range. The simulation results were obtained for a wire width of w = 5 μm, a structure height of h = 1 μm, and a gap width d between 100 μm and 100 nm.

Within the considered frequency range we observe a single peak corresponding to the fundamental SRR resonance. We find that the in-gap field strength is significantly enhanced and that it scales linearly with the ratio λ_r/d where λ_r denotes the wavelength at resonance. This is shown in Fig. 4. For the smallest considered gap width of d = 100 nm giant enhancement factors approaching 40000 are obtained which is more than an order of magnitude larger than the maximum field enhancement obtained with the nano-slit. This is explained by the resonant behavior where the oscillating currents excited in the SRR are much stronger than the non-resonant current flow induced on the metallic surface of the slit structures.

Linear Arrays of Nano-slits

The integrated nonlinear response from some material inside the nano-slit depends not only on the obtainable field strength but also on the volume comprising those high field strengths. Since for a single nano-slit the volume and, thus, the overall transmission is rather small the obvious approach is to arrange the slits into one-dimensional arrays with periodicity p as shown in Fig. 2(c). The total volume of the slits scales inversely with p for a given area of the sample. For small periodicities, however, neighboring slits start sharing the incident field energy so that, compared to single slits, the obtainable field enhancement is reduced.[14,15] The use of nano-slit arrays is consequently accompanied by a tradeoff between an increased volume and a decreased field enhancement.

For the realization of second or third order nonlinear processes the properties to be optimized are the ratios $F^2(p)/p$ and $F^3(p)/p$ respectively. Their dependence on the periodicity is shown in Fig. 5 for 40 nm wide slits at a frequency of 0.5 THz. The results indicate that optimal periodicity is around several tens of microns. The optimal value roughly scales with wavelength.

Square Arrays of SRRs

Due to the geometry of SRRs the possibility of increasing the nonlinear response by decreasing the periodicity is limited. We therefore increase the size of the high field strength volume by adding capacitive faces of length s to the gap as shown in the inset to Fig. 6.

As a consequence, the field enhancement decreases because the finite charge is distributed over a larger distance. As in the case of nano-slits this implies a tradeoff between an increased volume and a decreased field enhancement. For the realization of second or third order nonlinear effects again the volume of high field strength has to be optimized. Their dependence on the split length s is shown in Fig 6. Again we find optimal values on the order of several tens of microns.

Example Nonlinear Interaction

Two classes of questions arise immediately when claiming that such structures are useful for THz nonlinear interactions with matter.

The first is related to switching or steering applications where the high fields, which may be considered static on picosecond time scales, are used for example to influence charge transport within a molecule. In some cases quasi 'unipolar' THz pulses may be sufficient to influence the final state of matter. Such pulses can easily be generated since full control over THz pulse generation is state-of-the-art.[16] Other experiments may require some pump probe type measurements within the timeframe over which the THz field may be considered static. Since the geometry of the nano-structures may render the use of non-collinear pump and probe pulses impossible the most promising aspect for four wave mixing type detection seems to be phase cycling.[17]

The second question is whether the induced nonlinear response will be strong enough to be detected. For that purpose we have performed a numerical study where we investigate second harmonic generation (SHG) in the gap region of both nano-slit and the SRR structures filled with a nonlinear medium, here $LiTaO_3$.[18] Note, that the introduction of the dielectric substrate leads to a modification of the dielectric environment which results in an effectively smaller wavelength. According to our previous findings this leads to a decrease of the field enhancement as compared to a freestanding structure. The black curves in Fig. 7 show reference spectra of the pulse transmitted through a sample without any nano-structure. Here, the nonlinearity and the given field strength of the incident pulse is small so that hardly any SHG signal can be produced. The red curves, on the other hand, show the corresponding spectra of a pulse transmitted through the nano-slit (Fig. 7(a)) and the SRR arrays (Fig. 7(b)). The induced field enhancement leads to a much stronger SHG signal even though the nonlinear substrate material is only present below and not in the gap. The efficiency can be increased even further if the nonlinear material is directly inserted into the gaps. For that case, the obtained transmission spectra are shown as green curves in Fig. 7. Compared to air-filled gaps (red curves) the SHG signal is increased roughly twofold.

The SRRs were arranged in a two-dimensional array with periodicity 100 μm and gap width 1 μm. The exhibited behavior resembles the one of the nano-slits except that the amplitudes of the SHG spectra are much higher. The spectrum obtained for the SRR samples is somewhat narrower than the ideally doubled spectrum which is explained by the SRRs providing field enhancement only around resonance. The SHG spectrum of the slits, on the other hand, has approximately the same width as the ideal spectrum except that it is slightly red-shifted which is attributed to the field enhancement scaling with λ. The difference in the behavior exhibited by the two structures can consequently be used to realize either broadband or narrowband nonlinear interaction. The SRRs are thereby of particular interest because the narrowband conversion property allows for tunability of the nonlinearity by adjusting the structural parameters. Also note that even though the incident electric field is only 10 kV/cm the conversion efficiency (fields) for the slits and the SRRs is on the order 0.1% and 1%, respectively.

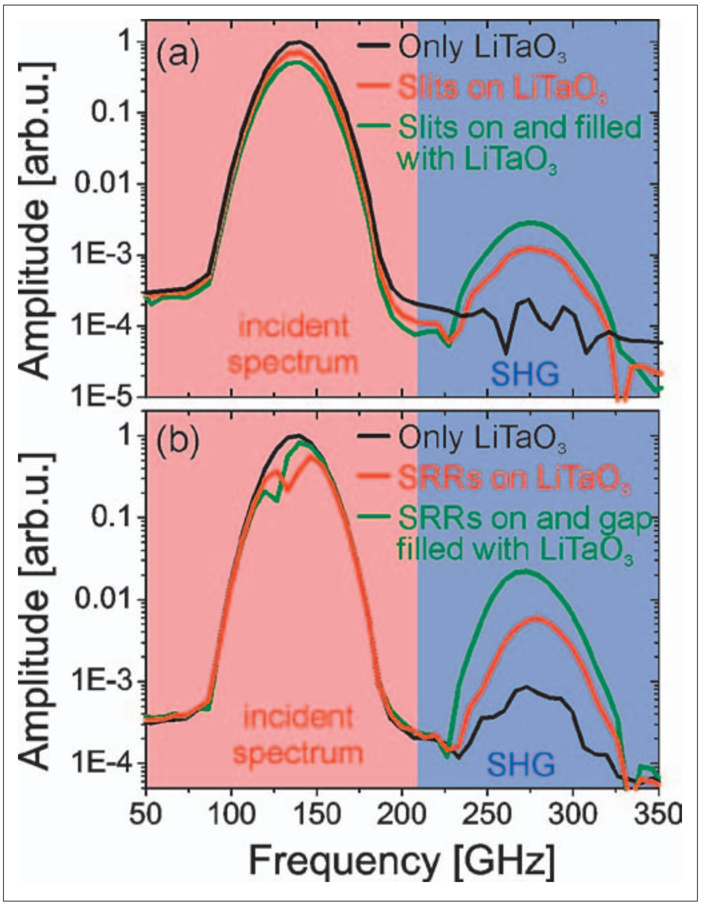

Fig. 7. (a) Transmission spectra of only the substrate, slit arrays with 40 nm slit width and a periodicity of 30 μm deposited on the substrate, and slit arrays where additionally the gap volume was filled. (b) As in (a) but for SRR arrays with d = 1 μm, L = 70 μm, and 100 μm periodicity.

Conclusion and Outlook

The goal of this brief overview was to show the prospects and limitations of using metallic nano-structures in THz switching applications or THz nonlinear spectroscopy. They also offer a new approach to light manipulation in THz domain and unique capability to engineer new photonics materials (so-called metamaterials) for THz applications. We believe that such structures will facilitate experiments not possible otherwise. We are convinced that these tools will also be useful for other projects within the MUST-NCCR.

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