

Terahertz nano-spectroscopy with resonant scattering probes

Tom Siday, Lucy L. Hale, Rodolfo I. Hermans and Oleg Mitrofanov

University College London, London, UK

We propose and demonstrate tunable resonant scattering probes for terahertz (THz) near-field microscopy, using sharp indium tips fabricated to the tine of a quartz tuning fork. We find the antenna resonance of the indium tips can be tuned by altering the tip length, which we support with numerical models. We also demonstrate the indium tips can provide nanoscale field confinement at the tip apex, with spatial resolution better than 100 nm.

I. INTRODUCTION

Terahertz nano-spectroscopy has the potential to provide unique insights into diverse physical phenomena, including dirac plasmons, intersubband polaritons, or even the ultrafast dynamics of excitons in two-dimensional materials. Whilst near-field microscopes with scattering probes can already reach the requisite nanoscale spatial resolution, the sensitivity remains too low to probe subtle variation in the local dielectric environment.

The efficiency in coupling to and scattering from the extremely subwavelength apex of a scattering tip is by far the greatest limitation to the sensitivity of near-field microscopy. Recently, it was shown that by designing a scattering probe to act like a resonant dipole antenna, coupling of far-field THz radiation to the apex can be significantly improved, enhancing the field at the tip apex[1,2]. However, these probes have not become popular with near-field researchers for two main reasons: firstly, it has proven difficult to produce resonant scattering probes cheaply and quickly, and second, there have been no direct measurements of the scattering efficiency of resonant tips. Here, we measure the efficiency of the entire

scattering process across the THz frequency range and directly demonstrate how the sensitivity of THz nano-spectroscopy can be dramatically increased using the principles of resonant scattering, using tips which can be fabricated without specialized equipment, and requiring minimal time investment.

II. RESULTS

Resonant scattering probes are fabricated from indium and mounted to the tine of a quartz tuning fork, which provide a compelling alternative to conventional atomic force microscope (AFM) cantilevers due to their high force sensitivity (Fig. 1(a)). To evaluate the scattering efficiency of the indium tips, we illuminate the tip with broadband terahertz pulses generated through optical rectification of in a ZnTe crystal[3]. By tapping the tip above the sample surface, and measuring at the first three harmonics of the tip tapping frequency (~ 32 kHz), we measure only the near-fields scattered from the tip apex. The THz fields scattered by the tip are then detected using a nanostructured photoconductive detector. Whilst the low numerical aperture of this detection scheme results in only small amounts of the scattered light being detected, this configuration enables direct probing of the THz fields incident on the scattering tip making it possible to qualitatively evaluate the scattering efficiency of the tip, by normalizing the scattered fields to the field incident on the tip.

We see a clear resonant peak in the scattering efficiency spectrum, for all three measured harmonics of the tip tapping frequency (Fig. 1(b), colored curves). The narrow spectral bandwidth, when compared to the incident THz spectrum (Fig. 1(b), black curve) provides direct evidence that the indium tips

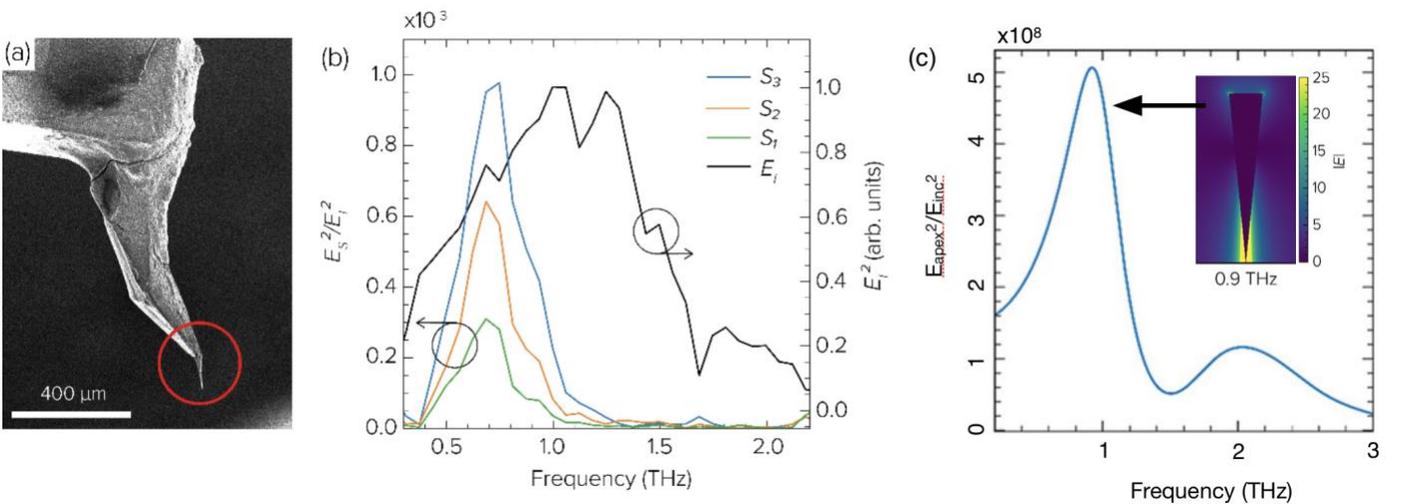


Fig. 1. Nano-spectroscopy with a resonant scattering probe. (a) Scanning electron micrograph (SEM) image of a 100 μm long indium tip, mounted to the tine of a quartz tuning fork. The indium tip is highlighted with a red circle. (b) Scattering efficiency of the resonant tip at the first three harmonics (S_1 , S_2 , S_3) of the tapping frequency (32 kHz). The spectra correspond to the tip shown in (a). The black curve shows the spectrum of the THz field incident on the indium tip. (c) Numerical model showing the resonant field enhancement at the apex of the scattering tip. Inset is the field distribution along the tip at the peaks of the $\lambda/2$ resonance (0.9 THz)[4].

resonantly scatter terahertz radiation, and can therefore dramatically improve the sensitivity of near-field microscopy. We further show that, through altering the geometry of the indium tip, the center frequency and bandwidth of the resonance can be shifted across the THz frequency range.

To confirm the origin of the observed spectral enhancement, we model the resonant scattering tips numerically, using a time domain solver (CST studio suite). When probing the field intensity at the tip apex, we see two clear resonant peaks in the spectrum (Fig. 1(c)). The lower frequency peak matches well with our experimentally extracted spectra for a 100 μm long tip, and we attribute this to a half-wave ($\lambda/2$) dipolar mode along the tip shaft.

In the numerical model, we also observe a peak near 2 THz, which cannot be extracted from our experimental results. We attribute this peak to a full wave (λ) resonance along the tip shaft, which, in the ideal dipole model, does not couple to the far-field. The finite geometry of our conical tip results in only minimal coupling of THz radiation to, and scattering from, the resonant tip. This, in combination with the lower SNR on our experiment at 2 THz, most likely results in this feature not being visible in Fig. 1(b).

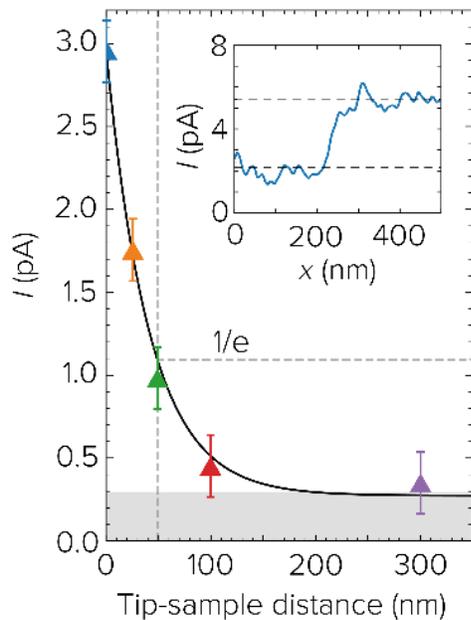


Fig. 2. Peak amplitude of the THz pulse scattered by the tip, for increasing tip-sample separation. Inset is the amplitude of the scattered field as the indium tip is scanned across a metal-dielectric interface[4].

The nanoscale THz field confinement at the apex of scattering tips is crucial for high-resolution near-field microscopy. To demonstrate nanoscale field confinement at the apex of the indium tips, we measure the peak THz electric field for increasing tip-sample separation (Fig. 2). We see the field decays by a factor $1/e$ in only 50 nm. We further confirm the nanoscale field confinement by scanning the tip across a metal-dielectric interface (Fig. 2, inset), demonstrating for the first time that indium resonant scattering probes can provide spatial resolution better than 100 nm, alongside the enhanced scattering efficiency.

III. SUMMARY

Resonant scattering probes present a promising approach towards achieving sensitive terahertz nano-spectroscopy. Here, we have provided a practical solution for the fabrication of resonant scattering probes, and have demonstrated that the resonant nature of these probes can be used to significantly increase the sensitivity of near-field microscopy.

REFERENCES

- [1] T. Siday *et al.*, “Resonant terahertz probes for near-field scattering microscopy,” *Opt. Express*, 25(22), 27874 (2017).
- [2] S. Mastel *et al.*, “Terahertz Nanofocusing with Cantilevered Terahertz-Resonant Antenna Tips,” *Nano Lett.*, 17(11), 6526–6533 (2017).
- [3] K. Moon *et al.*, “Quantitative coherent scattering spectra in apertureless terahertz pulse near-field microscopes,” *Appl. Phys. Lett.*, 101(1), 011109 (2012).
- [4] T. Siday *et al.*, “Resonance Enhanced Terahertz Nanoscopy Probes,” *ACS Photon.*, 7(3), 596-601 (2020).