HYPERSPECTRAL TIME-DOMAIN TERAHERTZ NANO-IMAGING

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Abstract: Terahertz (THz) near-field microscopy has wide and unprecedented application potential for nanoscale materials and photonic-device characterization. Here, we introduce hyperspectral THz nano-imaging, by combining scattering-type scanning near-field optical microscopy (s-SNOM) with THz time-domain spectroscopy (TDS). We describe the technical implementations that enabled this achievement and demonstrate its performance with a heterogeneously doped Si semiconductor sample. Specifically, we recorded a hyperspectral image of 40 by 20 pixel in 180 minutes, by measuring at each pixel a time domain spectrum covering the range from 0.4 to 1.8 THz. Fitting the spectra with a Drude model allows for measuring - noninvasively and without the need for Ohmic contacts - the local mobile carrier concentration of the differently doped Si areas with a spatial resolution of about ~70 nm. We envision wide application potential for THz hyperspectral nano-imaging, including nanoscale carrier profiling of industrial semiconductor structures or characterizing complex and correlated electron matter, as well as low dimensional (1D or 2D) materials.

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1. Introduction

Spectroscopy in the terahertz (THz) spectral range (~ 0.1 THz to 10 THz) enables the investigation of low-energy (meV range) properties of matter such as molecular rotations, lattice vibrations, spin waves and internal excitations of bound electron–hole pairs and conductivities [1-3]. Due to the long wavelength of THz radiation, however, the spatial resolution of conventional THz spectroscopy is limited by diffraction to about 150 μm (for a frequency of 1 THz, corresponding to a wavelength of 300 μm).

The diffraction limit can be circumvented by scattering-type scanning near-field optical microscope (s-SNOM). s-SNOM is based on atomic force microscope (AFM), where a metal tip is illuminated with focused laser beam. The tip acts as an antenna and focuses the illumination to a nanoscale near-field spot at the tip apex. Near-field interaction between tip and sample modifies the tip scattered light. Interferometric detection of the tip scattered light thus allows for probing local refractive and absorption properties of the sample, by recording amplitude and phase of the scattered field, respectively. The resolution is essentially determined by the tip radius, which is in the range of about 25 nm for commercial metal coated tips [4].

Owing to its unique high-resolution imaging and spectroscopy capability, s-SNOM has been applied to interrogate a wide range of material properties in the THz frequency range. It has been used with monochromatic sources (gas lasers, free electron lasers, QCL), which
allow for imaging and eventually spectroscopic imaging at selected wavelength [5-8]. s-SNOM has been also used with time domain spectroscopy (TDS) [9-11]. However, only point spectroscopy has been reported, i.e. spectra were acquired only at individual points. Imaging with TDS setups have been typically done with fixed delay line, where spectroscopy information is lost. Altogether, either monochromatic imaging or point spectroscopy has been reported [5-12]. Hyperspectral imaging, that is a recording of 2D array of TDS near-field spectra has been not achieved yet.

Here, by combining s-SNOM with THz-TDS, we demonstrate hyperspectral THz nano-imaging in the frequency range spanning 0.5 THz to 1.8 THz, by imaging charge carrier profiles of a heterogeneously doped Si semiconductor sample. Hyperspectral imaging was achieved by increasing the detector signal (i) via forward detection of the tip-scattered THz field and (ii) by placing the whole microscope setup into a nitrogen purge box to reduce THz absorption by water in the beam paths. The forward detection allows for operation without the need for a beam splitter, which increases the tip-scattered THz field reaching the detector. It is enabled by a dual-beam parabolic mirror design. Hyperspectral images were recorded by measuring at each pixel of a two-dimensional sample area a time domain spectrum covering 0.5 to 1.8 THz. By fitting individual THz TDS s-SNOM spectra with the help of the well-established finite dipole model of s-SNOM and a Drude model for doped Si, we measure the local mobile charge carrier density with a spatial resolution of about ~70 nm, noninvasively and without the need of any Ohmic contacts.

2. Result and discussion

![Fig. 1. Experimental setup and THz near field signal. (a) Schematic of THz-TDNS setup, (b) THz approach curve obtained with a commercial THz probe from Rocky mountain Tech (radius ~ 70 nm) taken on a Au surface, (c) far-field ($s_0$) and near-field spectra $s_n$ obtained by detector signal demodulation the n-th harmonic of the tip tapping frequency.](image-url)
The THz time-domain nanospectroscopy (referred to as THz-TDNS) setup is shown in Fig. 1.(a). It is a combination of a far-field broadband THz-TDS system (Menlo Co.) and a s-SNOM (Neaspec GmbH), see Fig. 4(a) for picture. THz radiation is generated by pumping a photoconductive semiconductor InAs antenna using a femtosecond laser (Menlo System) that has two fiber-coupled ports at $\lambda = 1560$ nm and an average output power of 70 mW. One arm of the excitation femtosecond laser with a larger energy than the band gap of the semiconductor generates photo-induced free carriers that are accelerated by a bias field. The accelerated charges generate electromagnetic radiation at THz frequencies (see Fig. S2 for far-field power spectrum)[13]. This THz radiation is focused onto a commercial AFM tip (Rocky mountain model: 25PtIr200B-H) using the parabolic mirror of the s-SNOM. Instead of detecting the backscattered field, as typically done in s-SNOM, here the tip-scattered THz field is measured in the forward direction. This detection scheme is enabled by the dual-beam parabolic mirror design of the Neaspec s-SNOM (illustrated in Fig. 1(a)). It avoids the use of a beamsplitter (required in backscattering operation) and thus increases the tip-scattered THz field reaching the detector. A photoconductive antenna similar to the emitter is used in reverse for detecting the forward-scattered THz pulses without the need for a cooled detector. An optical delay line is used to measure the THz electric field as a function of time, yielding interferograms. Amplitude ($s$) and phase ($\phi$) spectra of the tip-scattered field are obtained by Fourier transform of the complex-valued interferograms. All spectra were taken in a nitrogen atmosphere achieved by placing the THz-TDNS in a closed chamber filled with nitrogen (see Appendix 1, Fig. 4). Dry gas purging results in higher signal to noise ratio due to moisture removal.

The THz focus is diffraction limited (diameter $\sim 150 \mu m$) and thus illuminates a large part of the tip shaft and sample region, which produces a strong background signal that overwhelms the near-field signal resulting from tip-sample near-field interaction. The background signal is removed by demodulating the detector signal at a higher harmonic $n$ of the tip’s oscillation frequency $W$, as is routinely done in s-SNOM. The s-SNOM amplitude approach curves (Fig. 1.), acquired by measuring the demodulated detector signals as a function of the tip-sample distance at the n-th harmonic of the tip tapping frequency, illustrate the effect of signal demodulation. The $s_n$ signals are maximum at small tip-sample distances and decrease rapidly as the tip is pulled away from the sample. With increasing demodulation order the signal decay more rapidly with distance, as well known from s-SNOM [14]. For this work, approach curves were recorded routinely before THz spectra or THz near-field images were taken, in order to assure that background artifacts are eliminated. Normalized amplitude ($s_n$(sample)/$s_n$(reference)) and phase ($\phi_n$(sample)- $\phi_n$(reference)) THz near-field spectra are obtained by taking first reference spectra on a reference area (such as gold or silicon), followed by taking spectra at a desired position of the sample surface. As probes we used commercial metal tips of a tip length $L=80 \mu m$ and apex radius of $R \sim 70 \text{ nm}$ (Rocky mountain model: 25PtIr200B-H).

THz nano-spectroscopy enables noninvasive and contactless nanoscale carrier profiling of doped structures. To demonstrate this capability of our THz-TDNS setup, we studied a pre-characterized silicon static random access memory (SRAM) sample. The topography image (Fig(a)) shows several features between $\sim 40-420$ nm in height, which relate to implanted regions. According to the SRAM manufacturer, region D is the homogeneously p-doped substrate with a nominal hole density, $n_p = 2 \times 10^{16}$ cm$^{-3}$. Regions A, B and C are ion implanted. A is p-type with hole density, $n_p = 4 \times 10^{19}$ cm$^{-3}$, B and C are n-type with electron densities, $n_e = 1 \times 10^{17}$ cm$^{-3}$ and $n_e = 2 \times 10^{17}$ cm$^{-3}$, respectively. The near-field amplitude image $s_2$ shown in Fig(b) is taken by focusing the broadband THz radiation to the tip and raster scanning the sample. The scattered field from the tip-sample interface is collected using demodulation of the detector signal at higher harmonics of the tip resonance frequency. In Fig(b), the second harmonic amplitude images of the broadband scattered signal is shown and reveals the presence of regions with varying dopant levels. For spectroscopic characterization
of the different doping regions, the tip is illuminated with a THz radiation generated by pumping a photoconductive semiconductor InAs antenna as described above. A delay stage varies the arrival time of the signal with respect to the detection optical pulse. Scanning the delay line then allows to map out the electric field amplitude and phase of the THz waveform as a function of time. Following Fourier transformation, the signal collected by the detector is demodulated at a higher harmonic n (demodulation order, n=2 was used in this work) of the tip’s oscillation frequency. To obtain normalized nano-THz spectra, we took broadband spectra at different points on the sample (shown A-D in Fig(a) & (b)) and normalized them to a reference point shown as Ref. This reference point has a nominal electron density of $2 \times 10^{20}$ cm$^{-3}$, which implies that this region is metallic in the 0.4-1.8 THz spectral range. It thus can be used as a suitable reference to normalize the near-field spectra obtained on the regions A-D, analogously to normalizing to a Au or Si silicon surface as routinely done in nano-FTIR spectroscopy [15]. The experimental amplitude and phase point spectra are shown in Fig(c) & (e), respectively. We find that the average amplitude signal is different on the different sample regions, and generally tends to decrease with increasing frequency (Fig(c)). The phase spectra displayed in Fig(e) show also noticeably different spectra at the various doping regions. Particularly, the phase spectrum taken at point D shows decreasing phase with increasing frequency, while the spectra at points A-C exhibit an increasing phase with increasing frequency.
In order to analyze and interpret the THz TDNS spectra we performed model calculations using the well-established finite dipole model assuming a Drude-type dielectric function for the doped sample regions [16-18]. The model describes the tip by a metallic ellipsoid with a length of 600 nm and 40 nm apex radius, being illuminated at an angle of 60° relative to the tip axis. The tip-scattered field is given by $E_s = s e^{i\omega} \propto (1 + r_p)^2 \alpha_{eff} E_{inc}$, where $r_p$ is the far-field Fresnel reflection coefficient of the sample [18], $E_{inc}$ the incident electric field and $\alpha_{eff}$ the effective polarizability of the tip [19] that is governed by the near-field interaction between tip and sample (see Appendix 2 ). The local sample properties, i.e. the electron and...
hole densities $n_e$ and $n_p$, are included in $r_p$ via the dielectric function $\varepsilon$ of the sample. The presence of free carrier result in a Drude term given by the dielectric function $\varepsilon(n, \omega) = \varepsilon_{\infty} \left(1 - \frac{\omega_p^2}{\omega^2 + i\gamma \omega}\right)$ where $\varepsilon_{\infty} = 11.9$, $\omega_p = \sqrt{\frac{n\epsilon_0^2}{m^*}}$ is plasma frequency ($m^*$ is the effective mass of electron and $n$ is carrier concentration ($n_e$ or $n_p$)) and $\gamma$ is the damping constant, and is inversely proportional to the carrier concentration-dependent mobility $\mu(n)$ as $\gamma \sim 1/\mu (n_{\mu,e})$ \cite{20} (for more details on the parameters and equations used see Appendix 2). Our calculations include tip-sample distance modulation and higher-harmonic signal demodulation, yielding amplitude and phase spectra, $s_2(\omega)$ and phases $\phi_2(\omega)$ respectively \cite{18}. Figures 2(d) & (f) show the calculated near-field amplitude $s_2$ and phase $\phi_2$ spectra for the regions A-D, normalized to the n-doped Si region marked as Ref in Fig. 2(a). The carrier densities $n_e$ and $n_p$ were chosen such that best agreement with the experimental spectra (Figs. 2(c) & (e)) was achieved. They are shown in Table 1 and compared with the nominal values. We find reasonable agreement. Quantitative differences we explain by uncertainties in the model, which does not account for the realistic tip geometry.

The spectra and their variation with carrier concentration can be understood as follows. The tip-sample near-field interaction for a Drude-type semiconductor sample results in a step (respectively a weak resonance peak) in the amplitude spectrum near the plasma frequency $\omega_p$ \cite{21-23}, which is accompanied by a peak in the phase spectrum (Figs. 2(g) & (h)). The spectral behavior can be explained by a tip-induced plasmon resonance in the sample. Since the plasma frequency increases with carrier density, the amplitude step and phase peak shift to higher frequencies for higher carrier concentrations. This explains why the areas of higher carrier concentration (A, B, C) exhibit their phase maxima at frequencies larger than 1.8 THz, while area D (lowest doping concentration) shows a phase maximum at frequencies lower than 0.4 THz.

<table>
<thead>
<tr>
<th>Type</th>
<th>Nominal concentration (cm$^{-3}$)</th>
<th>Fitted concentration using finite dipole model</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$n^+$</td>
<td>$2 \times 10^{20}$</td>
</tr>
<tr>
<td>A</td>
<td>$p^+$</td>
<td>$4 \times 10^{19}$</td>
</tr>
<tr>
<td>B</td>
<td>$n^-$</td>
<td>$1 \times 10^{17}$</td>
</tr>
<tr>
<td>C</td>
<td>n-well</td>
<td>$2 \times 10^{17}$</td>
</tr>
<tr>
<td>D</td>
<td>p-epi</td>
<td>$2 \times 10^{16}$</td>
</tr>
</tbody>
</table>

In Fig. 3 we demonstrate the capability of the THz-TDNS setup for hyperspectral nano-imaging. To that end, we measured an array of 40 by 20 tip-enhanced TDS spectra at the n-p transition region C and D marked Fig. 3 (b) of the SRAM sample. After normalization of each spectrum to a reference spectrum, a hyperspectral amplitude (Fig. 3(c)) and phase (Fig. 3(d)) data cube A (x, y, $\omega$) is achieved, with x and y indicating the plane of the sample surface and the z-axis corresponding to the frequency axis (0.5 to 1.8 THz). The hyperspectral data shows the spectral dependent features of the n-p region discussed above (Fig. 2). Overall, larger amplitude, corresponding to higher carrier concentration, is observed on region C compared to region D. Monochromatic amplitude images – extracted from the hyperspectral data cube shows stronger amplitude signal differences of the n-p transition region at 0.6 THz compared to 1.6 THz (Figs. 3(e) & 3(g)). The phase hyperspectral data captures the complex spectral and spatial variation of the carrier density across the transition region C to D. As shown in the phase spectrum in Fig. 2(e), region D, compared to region C, displays higher phase for frequencies lower than 0.8 THz and weaker phase for frequencies larger than 0.8 THz. These phase spectral dependences are contained in the hyperspectral data and are displayed for clarity in the monochromatic phase images Fig. 3(f) (0.6 THz) and Fig. 3(h) (1.6 THz) that
are extracted from the hyperspectral data cube. Further, while the spatial boundary between region C and D is clearly visible in the amplitude hyperspectral data, the spatial phase boundary between region C and D is less clearly demarcated indicating possible carrier diffusion across the n-p interface as also observed recently [8]. Overall, these results demonstrate the sensitivity of THz-TDNS for spectral and spatial mapping of low carrier density locations, region C ($n_e = 2 \times 10^{17} \text{ cm}^{-3}$) and region D ($n_p = 2 \times 10^{16} \text{ cm}^{-3}$). The combined (spectral and spatial) set of information bundled in the data is unique to hyperspectral imaging that cannot be achieved by point spectra or individual monochromatic images [24]. We further note that while the THz images from these regions show large near-field contrasts, images taken at mid infrared (28 THz) or visible (474 THz) frequencies [see Appendix 1 Fig. 6] show no contrast between these regions, consistent with the calculation prediction shown in Figs. 2(g) & (h).

3. Conclusion

In summary, we introduced THz-TDNS (which combines s-SNOM with THz-TDS) for THz hyperspectral nano-imaging and demonstrated its capability for noninvasive, sensitive and contactless local mobile carrier concentration mapping at 70 nm spatial resolution. On average an integration time of 20 ms per pixel and 7 s per pixel was used for broadband and hyperspectral nano-imaging respectively. The THz-TDNS is designed to fit on kinematic mounts on either side of the AFM tip and is easily removal allowing for s-SNOM or tip-enhanced Raman or photoluminescence operation on the same neaSNOM platform. In addition, the THz-TDNS compact design avoids the need for a beam splitter (enabling
increased detected signal) and cooled detectors. We anticipate wide application potential for THz hyperspectral nanoimaging, including fast and non-invasive spectroscopic quantitative sampling of carrier densities in semiconductor nano-structures, complex and correlated oxides, low dimensional (1D or 2D) materials’ interfaces and boundaries at high spatial resolutions.

Appendix 1

Fig. 4. (a) Picture of THz-TDNS setup in a nitrogen purge box, (b) topography and (c) second harmonic broadband near field amplitude $s_2$ image of SRAM sample, (d) THz TDNS amplitude $s_2$ spectra measured at marked position of reference marked in (b) & (c) in (d) dry nitrogen environment and (e) in ambient.
Fig. 5. (a) Simulated THz amplitude $s_2$ and (b) phase $\varphi_2$ spectra of near-field contrast for regions A, B, C and D. Finite dipole model was used for the calculation resulting in p-doped Si fitted values $1 \times 10^{19}$, $3 \times 10^{17}$, $4 \times 10^{17}$, and $4 \times 10^{16}$ cm$^{-3}$ for A, B, C and D respectively, dash lines are simulated spectra without considering of reflection coefficient ($r_p = 0$).

Fig. 6. (a) Second harmonic near field amplitude $s_2$ image taken at mid IR wavelength, $\lambda = 10.5 \mu$m (28 THz), (b) forth harmonic near field amplitude $s_4$ image taken at visible wavelength, $\lambda = 632$ nm (474 THz).

Appendix 2

2.1 Fresnel reflection coefficient $r_p$ and effective polarizability $\alpha_{eff}$

Equations (1) and (2) are used to calculate Fresnel coefficient ($r_p$) for p doped silicon with thickness of $d_1$ [18]. Note that if $r_p$ is not taken into account in the calculation, simulated results do not give the best fit to experimental data (see Fig. 5 for comparison simulation with and without $r_p$). In these equations $\varepsilon_1$ is dielectric function of doped silicon, $\varepsilon_2$ is dielectric constant of the substrate which we assume $\varepsilon_2 = \varepsilon_{sl} = 11.9$ here, $\varepsilon_0 = 8.85 \times 10^{-12}$ Fm$^{-1}$ and $R_t$ is tip curvature radius which is 40 nm.

$$r_p(q, \omega) \approx \frac{\varepsilon_1 - \varepsilon_0}{\varepsilon_1 + \varepsilon_0}$$  \hspace{1cm} (1)

$$\varepsilon_* \approx \varepsilon_1 \frac{\varepsilon_2 + \varepsilon_1 \tanh qd_1}{\varepsilon_1 + \varepsilon_2 \tanh qd_1}, \quad q \approx 1/R_t$$ \hspace{1cm} (2)

Equations (3-9) gives us the effective polarizability [19]. $L$ is effective length of tip which considered 600 nm, $R_t$ is tip curvature radius, $C$ is the height-independent constant with $Q_0 = R_t^2 E_0$ which is the total amount of the polarization charge induced in tip in the absence of sample which is described in reference [17], $H$ is tip and sample distance.

$$\alpha_{eff} = C \left(1 + \frac{1}{2} f_0(H) g(H) + \frac{1}{2} f_0(H) g(H) \right)$$ \hspace{1cm} (3)

$$f_0(H) = \left( g - \frac{\frac{1}{2} W_0 + R_t}{2L} \right) \frac{\ln \frac{4L}{R_t}}{\ln \frac{4L}{R_t}}$$ \hspace{1cm} (4)

$$f(H) = \left( g - \frac{\frac{1}{2} W_0 + R_t}{2L} \right) \frac{\ln \frac{4L}{R_t}}{\ln \frac{4L}{R_t}}$$ \hspace{1cm} (5)

$$C = LQ_0/|E_0|$$ \hspace{1cm} (6)

$$\beta = \frac{\varepsilon_{dopedSi} - 1}{\varepsilon_{dopedSi} + 1}$$ \hspace{1cm} (7)

$$W_0 \approx 1.31 R_t L (L + 2R_t)$$ \hspace{1cm} (8)

$$W_t \approx R_t/2$$ \hspace{1cm} (9)

2.2 Dielectric function of p doped Si
Equations (10-12) are used to calculate the dielectric function in presence of carrier mobilities [22]. Here \( \varepsilon_{\infty} = 11.9 \) is the high-frequency dielectric constant of Si, \( \omega_p \) is plasma frequency and \( \gamma \) is damping. (In calculation we used the dielectric function of p doped Si for all four regions of A, B, C and D since the difference in dielectric function of p doped and n doped Si is very small and we assume that they are relatively the same.)

\[
\varepsilon_{doped\ Si}(\omega_{nd}) = \varepsilon_{\infty}(1 - \frac{\omega_p^2}{\omega^2 + i\omega\gamma})
\]

\[
\omega_p = \left(\frac{ne^2\times10^6}{\varepsilon_0\varepsilon_m m}\right)^{\frac{1}{2}} \times \frac{1}{200\pi\varepsilon_0}
\]

\[
\gamma = \frac{e^{100} \mu_{max}}{2\pi\varepsilon_0 m_0 \mu_0 (e^{pc/nd})^{1/2} (\mu_0 + (\mu))^{1/2}}
\]

Constant in the equations above are: \( \varepsilon_{\infty} = 11.9 \), \( \varepsilon_0 = 8.85 \times 10^{-12} \), \( e = 1.60218 \times 10^{-19} \), \( m = 0.37 \), \( m_0 = 0.91095 \times 10^{-30} \), \( c_0 = 2.99792 \times 10^8 \), \( \mu_0 = 44.9 \), \( \mu_1 = 7 \), \( \mu_{max} = 470.5 \), \( pc = 9.26 \times 10^{16} \), \( Cr = 2.23 \times 10^{17} \), \( Cs = 6.1 + 10^{20} \), \( \alpha = 0.779 \), \( \beta = 2 \).

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