Terahertz Near-field Imaging of Sub-wavelength Connected/Isolated Patches in Micro and Nanoscale: Experiment and Simulation of Optical and Electrical Properties

Changlin Wu,†‡ Guanjun You,¶ Chang Wang,†‡ and Juncheng Cao†‡

†Key Laboratory of Terahertz Solid-State Technology, Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences, 865 Changning Road, Shanghai 200050, China
‡Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China
¶Shanghai Key Lab of Modern Optical Systems, Terahertz Technology Innovation Research Institute, and Engineering Research Center of Optical Instrument and System, Ministry of Education, University of Shanghai for Science and Technology, Shanghai 200093, China

E-mail:

Tip apex source dipole radiating over samples.
Abstract

Drude model successfully quantifies the optical constants for bulk matters, but it is not suitable for sub-wavelength objects. In this paper, near-field optics microscopy and finite element simulation are used to study the gold patches of different connected states in the terahertz spectrum. Electron transportation is discovered in determining optical properties of micron conductors. As the sample size decreases, near-field interaction becomes more complex, and abnormal signal enhancement is observed on the substrate. With the help of simulation, the phenomenon of abnormal enhancement is discussed in detail, which lays the foundation for further experimental verification.

Introduction

Quantifying the optical property of terahertz (THz) sub-wavelength objects is one of the most difficult problems in the field of light-matter interaction. In most frequency spectrums, the optical properties are definite. Theoretical and experimental methods are used to extract from various materials. THz time-domain spectroscopy (THz-TDS) based on photoconducting dipole antennas was invented and used in the measurement of bulk materials. Because THz wave band is opaque to metals and water, strongly absorbed by some molecules and transparent to most dielectric materials. THz imaging and fingerprint technology are appealing to widely usage in the future. THz wave is sensitive to low-energy phenomena in bulk matters and molecules. These quantum phenomena can be simplified into classic oscillator models, like Drude model and Drude-Lorentz model.

Recent Studies found optical constants in THz range are different, of which 3 orders of magnitude between experimental to Drude model were reported. Size and shape of matter are vital in THz frequency as free electron gas assumption is invalid, and we have to analysis the inner electrical interaction. It is necessary to introduce the calculation method which adapts to the complex boundary conditions. And the sub-wavelength resolution optical equipment used to extract constants is needed, such as scattering-type scanning near-field
optics microscopy (s-SNOM) based on THz-TDS setup. Atomic force microscope (AFM) is the core device of s-SNOM. It produces strong electric enhancement at tip apex and radiates the near-field signal at each pixel to the detector. Sub 10 nm imaging resolution have been achieved by s-SNOM.

However, nonlinear effect of near-field imaging process makes optical properties not proportional to detected signal. The signal from each pixel is correlated to the background, and noise is inevitably introduced in demodulation. In s-SNOM experiment, it is a feasible alternative to measure and compare samples with known materials at the same time, and qualitatively reflecting the relative relation of their dielectric constants. But, as we mentioned before, those known materials may respond differently in the THz range and make the comparing results less reliable. When considering the THz-Nano matter interactions, it is essential to combine finite element method (FEM) simulation with experimental data. FEM simulation result is based on Maxwell’ equations, which ensure fixed fields strength ratio from different incident electric strength. Indeed, the experimental data of optical signals is also a relative value. To eliminate the difference between simulations and experiments, we need to modify simulation parameters until reaching experimental results.

Here, we carry out a fast FEM simulation method and transfer all pixels to near-field images (NFI), which can reduce the influence of computational error fluctuation on the result. On the high-resistance silicon substrate, there are connected patches and isolating patches. Micron and nanoscale samples fabricated by a focused ion beam (FIB) are measured via THz-TDS s-SNOM. Compared with simulated electric field distribution, the micron sample (1) has a good consistency, but the nanoscale sample (2) has some unexplained experimental phenomena. We speculate that the abnormal enhancement on the substrate is related to both surface roughness and signal enhancement caused by THz light irradiation. A proper experiment is needed to explain those phenomena.
Methods

Samples Fabrication

Focused Ion Beam Scanning Electron Microscopy (FIB-SEM, Helios G4 UX) is applied here to fabricate state-of-the-art nanometer 3D specimen and take immediate high-resolution SEM imaging. Compared with Electron-Beam Lithography and Extreme-Ultraviolet Lithography, FIB-SEM can directly etch down the metal film and keep a flat surface, while the metal lift-off technology will introduce a lot of unevenness and defects on the metal surface, including the sides. In this recipe, firstly vaporize a thin gold film (about 10 nm) on the high-resistance silicon substrate (resistivity greater than 20 kΩ cm, <111> orientation, thickness 512 µm). Secondly use FIB-SEM to fabricate it with in-situ SEM observation. The beam current is up to 60 pA, which takes only a few seconds to finish the etching process and maintain a relatively uniform surface. The pattern generator is used to control the Gallium ions’ acting range, which may lead to severe damage in the narrow ditch during the ions’ digging. The fabrication quality is checked by SEM and AFM. The resolution of the electron beam of Helios G4 UX is better than 1.2 nm and sufficient for the smallest structure in this experiment. The island growth and cross-section of the gold film are presented as insert images in Fig.1(a).

Experimental Setup

A commercial TDS s-SNOM (Neaspec, GmbH, Germany) is applied in this experiment to scan the THz near-field optical signal and AFM topography. Tunable gas laser (SIFIR-50, Coherent Inc., U.S.A) operating between 0.2 – 2.5 THz. The scattering signal is collected by THz bolometer (QMC Instruments Ltd., Cardiff, U.K.). A custom-designed probe with a length of up to 70 µm and tip apex radius of up to 40 nm is used to enhance the near-field effects. In this experiment, AFM is working in the tapping mode, which will periodically contact the sample surface. AFM tip scans (256 × 256) pixels area with the tip frequency
46.09 KHz and tapping amplitude at 175 nm.

**Simulation**

Commercial FEM software (COMSOL Multiphysics, RF module) is used to numerically solve the field distribution of real probe shape and sample structure. In order to reduce the calculation consumption of simulation, the metal probe shaft is hollowed out, impedance boundary or perfect electrical conductor (PEC) boundary conditions are used for metals, background electric field purely on Z-direction, and calculated results in frequency domain. The simulation domain is a sphere (filled with air $\varepsilon'_\text{air} = 1$) covered by a perfect matching layer. Using high-resistance silicon (real part of the refractive index $n'_\text{air} = 3.48$) only under the exposed substrate. Probe is a cone (shaft length is 70 $\mu$m) with a sphere (tip apex radius 40 nm) at the top to avoid discontinuities. In the full wave simulation, 2 THz p-polarized plane wave background field is applied to the 50 nm tip-sample junction. Scan all pixels of each sample and record the electric distribution near the tip-sample junction. Use internal self-grid division to mesh grids. Post process data with MATLAB into grayscale images.

**Sample-1**

The non-contact scanning in simulation is adopted in the 7 $\mu$m $\times$ 7 $\mu$m square area with step length 0.5 $\mu$m. Each pixel costs minutes to calculate on a common desktop computer and, in total, requires 40 hours (desktop:128G memory, i7-10700 CPU) to draw a simulation image. Simulate-image (SIMU) separately to ensure enough pixels and less computation time.

**Sample-2**

The non-contact scanning in simulation is adopted in the 2.2 $\mu$m $\times$ 2.2 $\mu$m square area with step length 0.1 $\mu$m. Each pixel costs minutes to calculate on a common desktop computer and, in total, requires 37 hours to draw a simulation image.
Results and Discusses

THz-TDS s-SNOM is a combined tool for acquiring optical signals and topography. THz pulses are focused on the tip-sample junction with p-polarized electric field, and dipole response is cultivated in the AFM probe. In a simplified consideration, the source dipole locates in the tip apex, moving and irradiates the underlining sample structure and reflects high-order signals (S₁, S₂, S₃, S₄, S₅) to the far-field. The samples are gold film on high-resistance silicon with structures fabricated by state-of-the-art FIB techniques. As depicted in Fig.1(a.), patches are dug up from the whole gold film and some patches will be connected to circumstance. The different electrical connected states will lead to strength varying of optical signal. On the Sample-1, gentle channels are down to tens of nanometers into the silicon substrate and forming two patches (see Fig.1(b.)). Optical signals S₁ to S₅ are collected at NFI.S₁-S₅. We compare the results of 3 points on NFI (averaged by 0.39 µm × 0.39 µm area). The connected patch (upper point) has the highest signal strength, and the isolated patch (bottom point) has an extremely weak reflected signal even compared with the Si-substrate (middle point). NFI.S₁-S₂ have robust values but a larger background basement. NFI.S₄-S₅ have nearly zero background basement but a significant noise disturbing. And the NFI.S₃ is a suitable candidate in our discussion. Although different orders of signals involve nonlinear optical processes, they are linearly correlated. As the background influence is equally applied to connected patches, isolated patches and silicon substrates. Assuming the incident background electrical field with frequency ω₀ is \( E_Z^{(0)}(\omega_0) \), the first-order signal \( S_1 = \alpha_1 E_Z^{(0)}(\omega_0 \pm f_0) \) generated by AFM probe modulation with a coefficient of \( \alpha_1 \) and tapping frequency \( f_0 \). Second-order and higher-order signals result from the mixing of different frequencies as \( S_n = \alpha_n E_Z^{(0)}(\omega_0 \pm n f_0) \), here the \( \alpha_n \) is the nonlinear modulation coefficient of different order signals. Similar results can be derived directly from analytical solutions. In this experiment, the signal strengths of Sample-1 have the following relationship

\[
2 \cdot E(\text{middle point}) = E(\text{upper point}) \times E(\text{bottom point})
\]
The side walls of Sample-1 are not steep ($\leq 400$ nm in lateral, $\leq 100$ nm in vertical), which is caused by the ions’ etching and result in material accumulation on the edges. Gallium mixed with Silicon oxide and even the Aurum will stack beside the channels as we can see from the profiles along the whole samples (see Fig.1 (b.)). Those hills-like structures weaken near-field signal due to the small area for interaction with the source dipole at the tip apex. However, the inclined slope will increase the contact area with the probe shaft, enlarge the plate area of the tip-sample capacitor, and radiate more high-order signals to the far field. As depicted in Fig.2 topography ridges extracted from (a.) and boundaries of high signal strength from (b.) are compared in (c.). The pattern of Fig.2(c.) gives an overview.
of the relationships between geometrical boundaries to sharply changed near-field signal boundaries. Even though the pattern is extracted from NFI.S1, it is highly fitted with other NFI results (see Fig.2(d.)). The optical edges are mostly encircled by the geometrical edges but some waving bottoms on the Si substrate also have great optical signals. The waving ranges of the structures is small (≤ 10 nm), which means that the topographical unevenness will have a decisive signal difference. This effect may be stronger than the varying caused by materials’ optical properties. The signal on the substrate near the connected patch is enhanced and we will discuss it further on Sample-2.

The connected patch has higher signal strength than the isolated one, which is solid evidence that the Drude model fails in sub-wavelength. The main reason for this phenomenon is the transport and balance of electrons, followed by the shape of conductors. FEM software COMSOL is introduced to investigate electrical property in simulation, and we lack proper experimental methods to verify it.

Simulation plays a vital role in the study of s-SNOM. Firstly, transform physical model (see Fig.1 (a.)) into meshing grids (see Fig.3 (a.)). Replace tapping process of AFM with a changing tip-sample distance in the frequency domain. The electric field generated from tip-sample junction is a nonlinear process, and it is inversely proportional to the cubic power of distance. However, photo-detector will average the radiation signals amongst tapping amplitudes and result in stable electrical signal when the amplitude is large enough. In this experiment, electrical signals from THz-TDS s-SNOM are almost unchanged when the amplitude is larger than 100 nm. So that a tip-sample distance of 50 nm is used in the simulation to replace the Fourier transform in the radiation detection process. This method will reduce the amount of calculation by dozens of times, which is the key to realizing the simulation of NFI.

External electric field is applied in Z-direction, which leads to the formation of source dipole $P_1$ at the tip apex. This source dipole will excite the charges on the probe shaft to produce dipole $P_2$ with opposite polarization. In this way, additional dipoles are formed
Figure 2: Extract the edges from AFM and SNOM images and compare them with NFI.S3. Scale bar 5 \( \mu \text{m} \).
along the entire probe axis. One of the results is that the probe lengths greatly influence the near-field strength $|E_Z|$, while the $|E_X|$ is relatively weak (see Fig.3(a.)). Drawing the electric field vectors on the sample surface of the isolated patch (Fig.3(b. and the connected patch (Fig.3(c. at position (0 $\mu$m, 0 $\mu$m) and (0 $\mu$m, 2.5 $\mu$m). All electric fields are perpendicular to the conductor surfaces of the probe and patches. The source dipole on the apex will have a strong electrical field pointing down the underneath structure. The isolated patch is electrically independent of the peripheral conductor film and dominant influenced by the tip apex. Directions of electric field vectors are different between the isolated and the connected patches. This phenomenon is caused by the free charge density $\rho_f$ on the conductive surface of the specimen. According to the boundary conditions of ideal electrical conductor and assuming the background electric field is point to Z-direction, there are $\hat{n} \times (\vec{E} - \vec{0}) = 0$ and $\hat{n} \bullet (\varepsilon\vec{E} - \vec{0}) = \rho_f$ on the surface. Regarding perpendicular conductor as the ground with free charge density $\rho_f^{(c)}$, which is equal to $\varepsilon_0 E_Z^{(0)}$ without source dipole excitation. Here $\varepsilon_0$ is the vacuum permittivity. The isolated patch is polarized by the source dipole on tip apex and results in negative free charge density $-\rho_f^{(i)}$. The sign is not important, because the frequency of THz wave is much higher than that of AFM tapping. All the detectable parameters are averaged through thousands of tapping cycles and related to the dependent variable $\rho_f$. Relative optical constants, like the ratio of permittivity $\varepsilon_{iso}/\varepsilon_{con}$, can be written in the expression

$$\frac{\varepsilon_{iso}}{\varepsilon_{con}} = \frac{\varepsilon_{patch}/\varepsilon_{Drude}}{|\rho_f^{(i)}|/|\rho_f^{(c)}|}$$

Near-field results from experiments and simulations have shown that electron transportation links electrical property to optical property. The free charge density excited by the source dipole will lead to a change of permittivity. The data between isolated patches and connected patches are given in the experiment, but their optical constants cannot be determined directly. On the contrary, FEM simulation starts with modelling, and the electric
field distribution results are obtained by setting optical constants. Comparing the experimental results (see Fig.1(c.)) and simulation (see Fig.3(d.)) can help us determine the optical constants. As depicted in Fig.3(d.), simulate normE of isolated and connected patches separately (SIMU.normE, normalized). By comparing the profiles data crossing centers, simulation highly agrees with experimental results of Eq. 1. Although our method is suitable for good metals (Au), bad metals and semiconductors are equally applicable. Firstly, approximate dielectric constant by Drude model, and then take near-field results and fit it with the aid of simulation to determine the optical constants of any kinds of sub-wavelength scale samples. The simulation also answers the problem of edge enhancement effects: more electrons are accumulated on the edges and result in higher signal strength. However, because the simulated EM waves are stored in the complex data form, the light-and-shade as shown in the figure is a problem that cannot be ignored.

Interaction between s-SNOM and nano-scale and micro-scale samples are different. Under the condition of FIB’s extreme precision, Sample-2 with 100 nm size and 10 nm etching depth is manufactured. Sample-2 consists of three groups of patches ranging from 100 nm to 300 nm and shapes of circles or squares. A group of square patches are connected outside, and their connecting lines are all equal in length. The tomography of Sample-2 has a small unevenness which is caused by the Si substrate (see Fig. 4(a.)). As the FIB etching process focuses the ion beam on a square frame, the etching intensity will increase when the focused area decreases. Therefore, when etching the gold film, it will inevitably lead to different etching depths. Unevenness on the silicon substrate will lead to patterns of signal enhancement. In this experiment, the optical signal on the substrate is much higher than that on the gold film. As depicted in Fig. 4(b.), signal strength on peripheral gold film is about 1.1 V, this result is higher than the previous one (see Fig. 1). Profiles 1-7 are acquired from NFI.S3, as gold film (profile 1), substrate (profile 3, 7) and patches (profile 2, 4, 5, 6). The recesses in the upper substrate have an extremely high signal strength reaching 3.0 V. And the signal strength of the upper left area is stronger than the lower right area. This strange
Figure 3: Finite Elements Simulation. (a) Models. (b) Electric field distribution of isolated patch. (c) Electric field distribution of connected patch. (d) Simulated images (grayscale normalized). Scale bar 1 μm.
phenomenon is not caused by the unevenness of the sample surface or incident angle of the THz wave, because profile 5 and profile 6 are highly coincident. The different shape patches in different positions have the same signal strength. However, as the patches’ size decreases, signal strength is non-uniform decrease and even equal to the larger patches’ results. This irregular strength of different patches can be verified by simulation (see Fig. 4(c.), profile 1). A simple explanation is that the comparable patches’ sizes (100 nm – 300 nm) to tip apex diameter (80 nm) will lead to free electron density change and antenna effects on the small metals. With the size shrinking, fewer free electrons are on the patches, while the electrons can absorb more light energy because of the higher circumference-area ratio of smaller patches.

As depicted in Fig. 4(c.), the SIMU.normE result is quite different from the experimental result. The undulation (Profile 7 in Fig.4(b.)) on Sample-2 may be an important factor of the enhancement effect as mentioned above (see Fig.2). However, under the same topography conditions, the signal around connected patches (Profile 4 in Fig.4(b.)) is stronger than that around isolated patches (Profile 5-6), which indicates effects from other factors. Study shown that THz wave can heat probe and then change carrier density of substrate. Based on the above facts, we propose the following assumptions:

1. The external electric field near the metals (tip apex, gold edges) is greatly enhanced. Electrons are heated and ejected into the silicon substrate. Heats to the substrate increase carrier density of substrate too.

2. Increasement of carrier concentration makes the silicon substrate more conductive. However, its conductivity must be weaker than that of the gold film, so the isolated patches (see Fig.4(b.)) show a higher signal ratio (Eq.2) than that in simulation (see Fig.4(c.)). And around the connected patches, carrier transportation is easier and results in larger signals.

3. Joule heat and carrier density will change substrate refractive index $n_s$. Sub-wavelength...
(aperture size $a$) metallic apertures resonant in the condition $\lambda/2 \approx n_s a$ also causes signal enhancement.

Those explanations need more parameters to take simulations and compare carefully with experiments.

Figure 4: NFI and SIMU images of Sample-2. (a.) AFM surface topography (256 × 256 pixels, profile 1,2). (b) S3 near-field imaging (256 × 256 pixels, profile 1-7). (c) Simulation results (step length 100 nm, Profile 1,2). Scale bar 1 µm.

Conclusions

We studied sub-wavelength objects in the THz range from experiments and simulation. THz-TDS SNOM provides us with near-field optical signal, which is related to the electric
distribution from FEM simulated. Different from Drude model results, optical properties of sub-wavelength gold-patches link to the confined electron transportation. Electron flows play a vital role in tip-sample interaction and influence far-field detected signals. 1.4×1.4 μm² gold patches (Sample-1) and a more elaborate Sample-2 were examined. Comparing simulated images with experimental results, we are further understanding the electrical properties of tip-sample junction. The signal strength of micro-scale patches is well determined by the free electron density, and proportional to the patches’ size. However, this phenomenon is more complicated when we conduct experiments on nano-scale samples (comparable to tip apex radius). First of all, the silicon substrate is abnormally enhanced, even with higher signals than the peripheral gold film. Secondly, the signals from different patches are not solely related to their size. We believe that these effects are mainly caused by the electron transportation caused by THz wave heating. We are looking for techniques to verify our conjecture and we believe the answer can help us further understand the THz-Nano matter interactions.

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References

(1) Palik, E. D. Handbook of optical constants of solids; Academic press, 1998; Vol. 3.

(2) Auston, D. H.; Nuss, M. C. Electrooptical generation and detection of femtosecond electrical transients. IEEE Journal of quantum electronics 1988, 24, 184–197.
(3) Smith, P. R.; Anoston, D. H.; Nuss, M. C. Subpicosecond photoconducting dipole antennas. *IEEE Journal of Quantum Electronics* **1988**, *24*, 255–260.

(4) Gallot, G.; Zhang, J.; McGowan, R.; Jeon, T.-I.; Grischkowsky, D. Measurements of the THz absorption and dispersion of ZnTe and their relevance to the electro-optic detection of THz radiation. *Applied Physics Letters* **1999**, *74*, 3450–3452.

(5) Cheon, H.; Yang, H.-j.; Lee, S.-H.; Kim, Y. A.; Son, J.-H. Terahertz molecular resonance of cancer DNA. *Scientific Reports* **2016**, *6*, 1–10.

(6) Hu, B. B.; Nuss, M. C. Imaging with terahertz waves. *Optics Letters* **1995**, *20*, 1716–1718.

(7) Wang, Q.; Xie, L.; Ying, Y. Overview of imaging methods based on terahertz time-domain spectroscopy. *Applied Spectroscopy Reviews* **2022**, *57*, 249–264.

(8) Qiu, F.; You, G.; Tan, Z.; Wan, W.; Wang, C.; Liu, X.; Chen, X.; Liu, R.; Tao, H.; Fu, Z., et al. A terahertz near-field nanoscopy revealing edge fringes with a fast and highly sensitive quantum-well photodetector. *Iscience* **2022**, *25*, 104637.

(9) Maissen, C.; Chen, S.; Nikulina, E.; Govyadinov, A.; Hillenbrand, R. Probes for ultra-sensitive THz nanoscopy. *Acs Photonics* **2019**, *6*, 1279–1288.

(10) Mastel, S.; Govyadinov, A. A.; Maissen, C.; Chuvilin, A.; Berger, A.; Hillenbrand, R. Understanding the image contrast of material boundaries in IR nanoscopy reaching 5 nm spatial resolution. *ACS Photonics* **2018**, *5*, 3372–3378.

(11) Walther, M.; Plochocka, P.; Fischer, B.; Helm, H.; Uhd Jepsen, P. Collective vibrational modes in biological molecules investigated by terahertz time-domain spectroscopy. *Biopolymers: Original Research on Biomolecules* **2002**, *67*, 310–313.

(12) Shen, J.; Zhu, Z.; Zhang, Z.; Guo, C.; Zhang, J.; Ren, G.; Chen, L.; Li, S.; Zhao, H. Ultra-broadband terahertz fingerprint spectrum of melatonin with vibrational mode
analysis. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy* 2021, **247**, 119141.

(13) Ordal, M. A.; Long, L.; Bell, R.; Bell, S.; Bell, R.; Alexander, R.; Ward, C. Optical properties of the metals al, co, cu, au, fe, pb, ni, pd, pt, ag, ti, and w in the infrared and far infrared. *Applied Optics* 1983, **22**, 1099–1119.

(14) Benavides-Cruz, M.; Calderón-Ramón, C.; Gomez-Aguilar, J.; Rodríguez-Achach, M.; Cruz-Orduña, I.; Laguna-Camacho, J.; Morales-Mendoza, L.; Enciso-Aguilar, M.; Pérez-Meana, H.; Escalante-Martínez, J., et al. Numerical simulation of metallic nanostructures interacting with electromagnetic fields using the Lorentz–Drude model and FDTD method. *International Journal of Modern Physics C* 2016, **27**, 1650043.

(15) Jeon, T.-I.; Kim, K.-J.; Kang, C.; Maeng, I. H.; Son, J.-H.; An, K. H.; Lee, J. Y.; Lee, Y. H. Optical and electrical properties of preferentially anisotropic single-walled carbon-nanotube films in terahertz region. *Journal of Applied Physics* 2004, **95**, 5736–5740.

(16) Pandey, S.; Gupta, B.; Chanana, A.; Nahata, A. Non-Drude like behaviour of metals in the terahertz spectral range. *Advances in Physics: X* 2016, **1**, 176–193.

(17) Park, D.; Choi, S.; Ahn, Y.; Rotermund, F.; Sohn, I.; Kang, C.; Jeong, M.; Kim, D. Terahertz near-field enhancement in narrow rectangular apertures on metal film. *Optics Express* 2009, **17**, 12493–12501.

(18) Chen, X.; Park, H.-R.; Pelton, M.; Piao, X.; Lindquist, N. C.; Im, H.; Kim, Y. J.; Ahn, J. S.; Ahn, K. J.; Park, N., et al. Atomic layer lithography of wafer-scale nanogap arrays for extreme confinement of electromagnetic waves. *Nature Communications* 2013, **4**, 1–7.

(19) Moon, K.; Park, H.; Kim, J.; Do, Y.; Lee, S.; Lee, G.; Kang, H.; Han, H. Subsurface
nanoimaging by broadband terahertz pulse near-field microscopy. *Nano Letters* 2015, 15, 549–552.

(20) Chen, X.; Liu, X.; Guo, X.; Chen, S.; Hu, H.; Nikulina, E.; Ye, X.; Yao, Z.; Bechtel, H. A.; Martin, M. C., et al. THz near-field imaging of extreme subwavelength metal structures. *ACS Photonics* 2020, 7, 687–694.

(21) Cocker, T.; Jelic, V.; Hillenbrand, R.; Hegmann, F. Nanoscale terahertz scanning probe microscopy. *Nature Photonics* 2021, 15, 558–569.

(22) Hillenbrand, R.; Taubner, T.; Keilmann, F. Phonon-enhanced light–matter interaction at the nanometre scale. *Nature* 2002, 418, 159–162.

(23) Cvitkovic, A.; Ocelic, N.; Hillenbrand, R. Analytical model for quantitative prediction of material contrasts in scattering-type near-field optical microscopy. *Optics Express* 2007, 15, 8550–8565.

(24) Knoll, B.; Keilmann, F. Enhanced dielectric contrast in scattering-type scanning near-field optical microscopy. *Optics Communications* 2000, 182, 321–328.

(25) Chen, X.; Hu, D.; Mescall, R.; You, G.; Basov, D.; Dai, Q.; Liu, M. Modern scattering-type scanning near-field optical microscopy for advanced material research. *Advanced Materials* 2019, 31, 1804774.

(26) Pizzuto, A.; Chen, X.; Hu, H.; Dai, Q.; Liu, M.; Mittleman, D. M. Anomalous contrast in broadband THz near-field imaging of gold microstructures. *Optics Express* 2021, 29, 15190–15198.

(27) Zhang, Z.; Hu, M.; Zhang, X.; Wang, Y.; Zhang, T.; Xu, X.; Zhao, T.; Wu, Z.; Zhong, R.; Liu, D., et al. Direct observation of tip-gap interactions in THz scattering-type scanning near-field optical microscopy. *Applied Physics Express* 2021, 14, 102004.
(28) Huth, F.; Chuvilin, A.; Schnell, M.; Amenabar, I.; Krutokhvostov, R.; Lopatin, S.; Hillebrand, R. Resonant antenna probes for tip-enhanced infrared near-field microscopy. 
Nano Letters 2013, 13, 1065–1072.

(29) Chen, X.; Lo, C. F. B.; Zheng, W.; Hu, H.; Dai, Q.; Liu, M. Rigorous numerical modeling of scattering-type scanning near-field optical microscopy and spectroscopy. 
Applied Physics Letters 2017, 111, 223110.

(30) McArdle, P.; Lahneman, D.; Biswas, A.; Keilmann, F.; Qazilbash, M. Near-field infrared nanospectroscopy of surface phonon-polariton resonances. Physical Review Research 2020, 2, 023272.

(31) Chen, X.; Yao, Z.; Xu, S.; McLeod, A. S.; Gilbert Corder, S. N.; Zhao, Y.; Tsuneto, M.; Bechtel, H. A.; Martin, M. C.; Carr, G. L., et al. Hybrid machine learning for scanning near-field optical spectroscopy. ACS Photonics 2021, 8, 2987–2996.

(32) McLeod, A. S.; Kelly, P.; Goldflam, M.; Gainsforth, Z.; Westphal, A. J.; Dominguez, G.; Thiemens, M. H.; Fogler, M. M.; Basov, D. Model for quantitative tip-enhanced spectroscopy and the extraction of nanoscale-resolved optical constants. Physical Review B 2014, 90, 085136.

(33) Mooshammer, F.; Huber, M. A.; Sandner, F.; Plankl, M.; Zizlsperger, M.; Huber, R. Quantifying nanoscale electromagnetic fields in near-field microscopy by Fourier demodulation analysis. Acs Photonics 2020, 7, 344–351.

(34) Feres, F. H.; Mayer, R. A.; Wehmeier, L.; Maia, F. C.; Viana, E.; Malachias, A.; Bechtel, H. A.; Klopf, J. M.; Eng, L. M.; Kehr, S. C., et al. Sub-diffractional cavity modes of terahertz hyperbolic phonon polaritons in tin oxide. Nature Communications 2021, 12, 1–9.

(35) Seo, M.; Park, H.; Koo, S.; Park, D.; Kang, J.; Suwal, O.; Choi, S.; Planken, P.;
Park, G.; Park, N., et al. Terahertz field enhancement by a metallic nano slit operating beyond the skin-depth limit. *Nature Photonics* 2009, 3, 152–156.

(36) Wiecha, M. M.; Kapoor, R.; Roskos, H. G. Terahertz scattering-type near-field microscopy quantitatively determines the conductivity and charge carrier density of optically doped and impurity-doped silicon. *APL Photonics* 2021, 6, 126108.

(37) Kang, J.; Choe, J.-H.; Kim, D.; Park, Q.-H. Substrate effect on aperture resonances in a thin metal film. *Optics Express* 2009, 17, 15652–15658.

(38) Adak, S.; Tripathi, L. N. Nanoantenna enhanced terahertz interaction of biomolecules. *Analyst* 2019, 144, 6172–6192.