Phase-resolved Detection and Control of Ultrabroadband THz Pulses coupled to a Scanning Tunneling Microscope Junction

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Coupling phase-stable single-cycle terahertz (THz) pulses to scanning tunneling microscope (STM) junctions enables spatio-temporal imaging with femtosecond temporal and Ångstrom spatial resolution. The time resolution achieved in such THz-gated STM is ultimately limited by the sub-cycle temporal variation of the tip-enhanced THz field acting as an ultrafast voltage pulse, and hence by the ability to feed high-frequency, broadband THz pulses into the junction. Here, we report on the coupling of ultrabroadband (1-30 THz) single-cycle THz pulses from a spintronic THz emitter (STE) into a metallic STM junction. We demonstrate broadband phase resolved detection of the tip-enhanced THz waveform via THz-field-induced modulation of ultrafast photocurrents across the junction. Comparison to the unperturbed far-field THz waveform reveals the antenna response of the STM tip. Despite tip-induced low-pass filtering, frequencies up to 15 THz can be detected in the enhanced near-field, resulting in THz transients with a half-cycle period of 115 fs. We further demonstrate versatile phase and polarity control of the THz waveform depending on the STE excitation conditions and magnetization, and show that up to 2 Volts THz bias at 1 MHz repetition rate can be achieved in the current setup. Finally, we find a nearly constant THz voltage and waveform over a wide range of tip-sample distances, which by comparison to numerical simulations confirms the quasi-static nature of the THz pulses. Our results demonstrate the suitability of spintronic THz emitters for ultrafast THz-STM and provide insight into the femtosecond response of defined nanoscale junctions.

KEYWORDS: scanning tunneling microscopy, THz near-field sampling, spintronic THz emitter, broadband THz pulses, tip antenna response
THz-gated scanning tunneling microscopy (THz-STM) combines Ångstrom spatial with femtosecond temporal resolution, which has been impressively demonstrated on single molecules\textsuperscript{1} and semiconductor surfaces\textsuperscript{2}. Following the original idea of junction-mixing STM\textsuperscript{3,4}, the concept of THz-STM is based on the rectifying nature of an STM junction exhibiting nonlinear I-V characteristics, leading to a net DC current upon modulation of the junction bias with ultrafast voltage pulses. Adapting this concept, THz-STM utilizes ‘wireless’ free-space coupling of ultrafast voltage pulses to the STM by illumination with coherent broadband THz radiation\textsuperscript{1,2,5–8}. The STM tip hereby acts as a broadband antenna strongly enhancing the quasi-static THz electric field, allowing for the application of large sub-picosecond bias pulses at moderate incident THz field strength. Optimizing THz-STM operation requires precise knowledge of the THz voltage transient across the junction and hence broadband characterization of the tip antenna response in the STM environment.

The coupling of THz radiation to scanning probe tips has been widely studied in the context of THz scanning near-field optical microscopy\textsuperscript{9–14} (THz-SNOM). It is known that metallic tips act as long wire antennas\textsuperscript{10,11}, which low-pass filter the broadband incident THz radiation\textsuperscript{9} and exhibit highly directionally emitting and receiving properties\textsuperscript{11}. The STM tip-enhanced THz waveform will, thus, differ considerably from the incident THz waveform, depending on the specific STM geometry, the mesoscopic shape of the tip wire, as well as the incident THz bandwidth. Yet, detailed experimental characterization of the STM tip antenna properties and its effect on the THz waveform inside the STM environment remain scarce\textsuperscript{7,8}. In particular, understanding the antenna response of the STM tip over a broad frequency range exceeding 10 THz will be crucial to extend THz-STM towards high-
frequency single-cycle THz pulses up to the multi-THz and mid-infrared regime\textsuperscript{15,16}, potentially increasing the time resolution achievable in THz-STM.

Recently, ultrabroadband single-cycle THz pulses were successfully generated from a metallic spintronic THz emitter (STE) with spectra covering the frequency range up to 30 THz without gap\textsuperscript{17,18}. The spectral bandwidth of the STE output is determined by the duration of the incident pump pulse and the resulting carrier dynamics in the STE\textsuperscript{19}. In addition to its extremely large bandwidth and fast THz transients, the STE exhibits several advantages such as convenient THz polarity switching and polarization control via the STE magnetization, flexibility regarding pump photon energy, pulse duration, and excitation geometry, its easy handling as well as low cost\textsuperscript{20}. Although the conversion efficiency of the STE is considerably lower compared to standard THz sources such as LiNbO\textsubscript{3}\textsuperscript{21,22} or photoconductive antennas\textsuperscript{23,24}, its high beam quality in combination with high THz bandwidth allows for tighter focusing, facilitating peak field strength of 300 kV/cm at few mJ pump pulse energies\textsuperscript{18}. Hence, high electric field strengths are achieved at comparably low THz power, making the STE an attractive THz source in particular for field-driven applications requiring high repetition rates such as THz-STM.

Here, we report on the experimental characterization of ultrabroadband THz pulses coupled to a metallic STM junction in the frequency range from 1 to 30 THz at 1 MHz repetition rate. To characterize the bandwidth, phase and voltage amplitude of the tip-enhanced THz field we sample its waveform directly in the time domain by THz-induced modulation of ultrafast photocurrents excited with near-infrared (NIR) femtosecond laser pulses in the tip-sample junction\textsuperscript{8,25}, as sketched in \textbf{Figures 1a)} and \textbf{1b)}. By comparison to the free-space THz waveform measured via electro-optic sampling (EOS), we can experimentally determine the
receiving antenna response of the STM tip. Special care has to be taken to obtain the unperturbed tip-enhanced THz waveform without distortion by photoelectron dynamics, which is particularly crucial at the broad bandwidth employed here. We demonstrate versatile phase and polarity control of the THz waveform in the STM junction via the STE excitation conditions. Peak THz voltages of several Volts can be reached in the STM junction at moderate pump pulse energies of a few microjoules facilitating repetition rates in the MHz regime with fiber-based laser sources. Finally, we analyze the distance dependence from the tunneling regime to the µm range, revealing a nearly constant THz voltage in line with the quasi-static nature of electromagnetic radiation when applied to sub-wavelength dimensions. Our results demonstrate the suitability of spintronic THz emitters as broadband source for the application of ultrafast voltage transients in STM and highlight the importance of the tip antenna response that exhibits significant low-pass filtering and reduction of the THz bandwidth available for THz-STM operation. Aiming at even higher time resolution in THz-STM, thus, requires strategies for selective enhancement of elevated THz frequencies in the THz-biased STM junction.

**Experimental details.** The STE is excited with broadband 800 nm NIR pulses of 8 fs duration under normal incidence, leading to THz pulse emission collinear with the NIR laser beam. The STE magnetization is controlled by a permanent magnet. The STE position along the focused NIR pump beam can be set by a translation stage, thereby enabling waveform
control of the THz pulses as discussed below. The THz pulses are focused onto the STM tip via an off-axis parabolic mirror (35 mm focal length) integrated on the STM platform inside the ultrahigh vacuum (UHV) chamber. A second parabolic mirror is used to focus 8 fs NIR laser pulses into the STM (NIR spot size ~ 6 µm) for photoexcitation of the junction at a variable time delay $\Delta t$ compared to the arrival time of the THz pulses. Both THz and NIR

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{(a) Experimental scheme and (b) junction diagram for sampling of the THz voltage transient applied to the STM. The THz electric field acts as an instantaneous bias which modulates the photocurrent generated by 8 fs NIR laser pulses. Depending on the barrier height and distance, the photocurrent originates from above-barrier photoemission and/or photo-assisted tunneling through the barrier. (c) Laser-induced photocurrent-voltage characteristics and (d) tip-enhanced THz waveforms sampled at 8 V DC bias ($d = 1 \mu m$, $E_{\text{p,STE}} = 0.6 \mu J$) for positive and negative THz polarity (STE magnetization $M^+/M^-$). The THz voltage (left y-axis) is retrieved from the THz-induced photocurrent change (right y-axis) via the linear I-V slope (green dashed line in c). The power law scaling of the photocurrent (inset in (c), $U_{\text{DC}} = 8$ V) reveals operation in the multiphoton photoemission regime.}
\end{figure}
pulses are polarized along the tip axis. All experiments are performed at room temperature and under UHV (pressure 1e-10 mbar) conditions.

**THz-induced photocurrent modulation.** Figure 1c) shows the photocurrent-voltage characteristics of the photoexcited junction at a tip-sample distance of 1 µm in the absence of the THz pulse. As the NIR focus is larger than the tip-sample distance, photoelectrons are excited both from tip and sample and the photocurrent $I_{ph}$ reverses sign at small negative DC bias. At large positive (negative) DC bias, as sketched in Figure 1b), the photocurrent is dominated by pure photoelectron emission from the tip (sample), whereas in the low bias regime, it is composed of both tip and sample photoelectrons depending on their respective energy distributions. At large distances, photoemission occurs either in the multiphoton regime\textsuperscript{26–28} or optical field-driven regime\textsuperscript{29–31} depending on the applied NIR field strength and classified by the Keldysh parameter\textsuperscript{32}. With our experimental parameters, we obtain a Keldysh parameter in the range of $\gamma \sim 10$-20. Hence, we clearly operate in the multiphoton photoemission regime, as confirmed by the scaling of the power dependence of the photocurrent shown in the inset in Figure 1c). At high DC bias and, in particular, at short distances, photo-assisted tunneling through the narrowed barrier\textsuperscript{33} may also contribute to or even dominate the photocurrent.

Applying a THz pulse to the STM junction in addition to the static DC bias leads to a change in the laser-induced photocurrent due to the THz field\textsuperscript{8,25}. As the time scale of photoemission is given by the temporal envelope of the 8 fs NIR laser pulse and occurs thus nearly instantaneous on the time scale of the THz pulse, the photoemission process is sensitive only to the *instantaneous* THz field at a given NIR-to-THz pulse time delay $\Delta t$. One may, thus,
make the quasi-static approximation which assumes that the THz-induced change in photocurrent will be determined by the local slope of the static $I_{ph}$-V characteristics, as sketched in Figure 1c). As will be demonstrated below, our measurements corroborate this assumption provided that non-instantaneous effects are eliminated. In the case of a linear $I_{ph}$-V curve with local slope $m_{I-V}$, as obtained at high positive bias in Figure 1c), the instantaneous THz voltage $U_{THz}(\Delta t)$ can be obtained from the THz-induced change in photocurrent $\Delta I_{THz}$ simply by $U_{THz}(\Delta t) = \Delta I_{THz}(\Delta t)/m_{I-V}$. Varying the time delay $\Delta t$, thus, directly yields the calibrated THz voltage transient as plotted in Figure 1d), measured at 8 V DC bias and 1 µm gap distance. Reversing the sign of the STE magnetization flips the THz polarity, and we find that the tip-enhanced THz waveform also reverses sign while maintaining its overall shape. Besides demonstrating this strikingly simple THz bias control in the STM provided by the STE magnetization, this finding confirms the interpretation of the employed sampling technique.

At this point, it is important to emphasize that non-instantaneous effects such as photoelectron propagation in the junction and long lifetimes of hot electrons inside the photoexcited tip and sample surface could alter the measured waveform and, thus, have to be ruled out. The lifetime of hot electrons in metals is in the range of few fs to several tens of fs depending on the sampled energy window and temperature. At far gap distances, lifetime effects can be neglected because the photoelectrons are emitted predominantly above the barrier, and tunneling can occur only at the very barrier top, where extremely short lifetimes of few fs are expected. Furthermore, THz streaking of photoelectrons back into the tip/sample could also result in a modified waveform, but can be precluded by operating
at high DC bias and low THz field strength (smaller than the DC field). The latter is only strictly given in the single electron regime, where the electron trajectories are solely determined by the combined DC and THz field. At larger photocurrents with many hundreds to thousands of electrons per pulse, space charge dynamics has to be considered and can significantly alter the measured THz waveform by acting as an effective low-pass filter. Hence, care has to be taken to operate in a low-excitation limit at small photocurrents. We find that 10 to few 100 electrons per pulse are usually sufficiently low to neglect space charge effects (see Supporting Information for more detailed information). It should be noted, however, that the critical photocurrent depends on the net accelerating forces acting on the photoelectrons, and, thus, on the DC field as well as on the space charge density in the junction, i.e., on the effective photoemission area and tip-sample distance. To ensure instantaneous THz near-field sampling within the quasi-static approximation, we thus, verify

Figure 2: Comparison of (a) incident THz electric field and (b) tip-enhanced THz voltage transient in the STM junction ($U_{DC} = 8$ V, $d = 1 \mu m$, $F_{p,STE} = 0.6 \mu J$). (c) THz amplitude spectra of the waveforms of (a) and (b), revealing strong low-pass filtering from the tip, which is characterized by (d) the measured receiving antenna transfer function $H(\omega)$. The dark green lines in (d) show the response of the fitted antenna model with $R = 102 \Omega$, $L = 0.96$ pH and $C = 4.5$ fF, yielding a resonance frequency of 0.25 THz.
that the sampled THz waveform is insensitive to the THz polarity, incident THz field strength, DC bias and tip-sample distance (details are discussed in the Supporting Information and in Figure 5 below).

**Tip-antenna response function.** Having established a reliable method to sample the THz waveform inside the STM junction now allows for the characterization of the tip antenna response by comparison of the unperturbed incident THz field and the received THz voltage transient. **Figure 2a** shows the THz electric field \( E_{in} \) obtained by electro-optic sampling using a 300 µm thick ZnTe(110) detection crystal in an identical reference beam path and after deconvolution with the detector response\(^{36} \). The corresponding THz voltage transient in the STM is shown in **Figure 2b**. The low-pass filter effect of the tip antenna becomes immediately obvious in the time domain data, where a half-cycle peak-to-peak separation of 115 fs is attained in the tip-enhanced waveform, noting the maximum speed at which the THz bias can be reversed in the STM. In the time domain, the THz near-field in the STM junction \( E_{STM}(t) \) is given by convolution of the incident THz field \( E_{in}(t) \) with the tip impulse response. More convenient access to the tip antenna response is obtained by Fourier transformation of the time domain data into the frequency domain, where the tip-enhanced THz field is simply the product of the incident THz field and the complex tip transfer function \( H(\omega) \). Equivalent to a receiving antenna’s output voltage and following wideband antenna theory,\(^{37} \) we can also use the experimentally accessible frequency-dependent THz voltage \( U_{THz}(\omega) \) instead of the tip-enhanced THz field and relate it to the incident THz electric field \( E_{in}(\omega) \) by

\[
U_{THz}(\omega) = H(\omega) E_{in}(\omega).
\]  

(1)
Here, $H(\omega)$ is now the receiving antenna transfer function with units in meters. It also depends on the angle $\theta$ of incidence of the THz beam. The Fourier spectra of the unperturbed and tip-enhanced waveforms are plotted in Figure 2c), revealing a pronounced bandwidth reduction and simultaneous shift of the tip-enhanced THz spectrum to lower frequencies. The tip transfer function $H(\omega)$ is then simply obtained by dividing the two complex Fourier spectra, and its amplitude and phase are plotted Fig. 2d). The amplitude $|H|$ reflects the coupling efficiency of the tip antenna at a specific THz frequency. To estimate the resonance frequency of the tip antenna, the measured transfer function is fit using a simple model which treats the tip antenna as an $RLC$ circuit with resistance $R$, capacitance $C$ and inductance $L$ connected in series.9,25 (see Supporting Information for more details). The best fit with $R = 102 \ \Omega$, $L = 0.96 \ \mu\text{H}$ and $C = 4.46 \ \text{fF}$ yields a resonance frequency of $f_0 = \frac{1}{(2\pi \sqrt{L'C})} =$
0.25 THz. The spectral phase of the measured transfer function approximately follows the phase of the modelled resonance circuit, which is flat for frequencies above 1 THz and exhibits a $180^\circ$ phase shift at the resonance. Hence, no significant distortion (chirp) of the THz pulse is introduced by the tip for frequencies $> 1$ THz due to the absence of group delay dispersion in this frequency range. Tuning the tip resonance frequency and its transfer function via resonant tip shaping should, thus, allow for control of the tip-enhanced THz near-field waveform in the STM.

**THz pulse control.** We now use our previous results to demonstrate control of the THz voltage waveform applied to the STM junction. We start by introducing a phase shift to the incident THz field by moving the STE inside the convergent beam of the NIR pump laser, as sketched in Figure 3a). In such a curved-wavefront excitation scheme the local radius of curvature of the NIR beam is imprinted on the generated THz field.\(^{23}\) Upon propagation to the far field the emitted THz pulse thus acquires a frequency-dependent Gouy phase shift that depends on the STE position. In particular, an intermediate THz focus is generated when placing the STE in the convergent NIR beam.\(^ {23}\) As seen in the top panel in Figure 3b), we observe a transformation from a rather symmetric to a more asymmetric pulse shape of the tip-enhanced THz waveform when moving the STE further away from the NIR focus.

To model this behavior, we apply the measured tip transfer function to t phase-shifted incident THz fields and compare the reconstructed antenna-enhanced waveforms to the corresponding THz waveforms measured in the STM. Specifically, we multiply the complex Fourier spectra $E_{in}(\omega)$ of the deconvoluted THz waveforms obtained from EOS (see Figure S3 in the Supporting Information) with $H(\omega)$ to obtain the calculated Fourier spectra
$U_{\text{rec}}(\omega)$ of the THz voltage received by the tip antenna. Inverse Fourier transformation of $U_{\text{rec}}(\omega)$ then yields the antenna-enhanced THz waveforms plotted in the bottom panel in Figure 3b). Comparison of the waveforms for three different STE positions reveals that the small phase shifts apparent in the measured THz near-field waveforms are clearly reproduced by the respective calculated waveforms, demonstrating the validity of the experimentally obtained tip transfer function.

Further control of the THz waveform via the Gouy phase is demonstrated in Figure 3c), where tip-enhanced THz waveforms are plotted for different positions $z_{\text{rec}}$ of the THz recollimation mirror with the STE positioned 1 mm in front of the NIR focus. Moving this mirror alters the THz beam divergence and, thus, shifts the THz focal plane with respect to the tip position, changing the local phase of the focused THz beam seen by the STM tip. This procedure allows for continuous control of the THz waveform from a symmetric to an asymmetric pulse shape. The corresponding peak THz voltages vary according to the

Figure 4: (a) Peak THz voltage vs STE pump pulse energy for different positions of the STE ($\Delta z_{\text{STE}}$) in front of the NIR focus. The THz voltage scales sublinear with the pump photon energy due to thermal saturation depending on the NIR spot size and hence STE position. Power exponents $n$ are obtained from the fits (solid curves). At very small spot sizes close to the NIR focus ablation occurs and prevents excitation at high pump energies in the current setup. (b) Peak THz voltage as a function of STE position ($E_{\text{p,STE}} = 1.2 \, \mu J$). The black dashed line shows the NIR pump spot size vs STE position (right y-axis). ($U_{\text{DC}} = 8 \, V, d = 1 \, \mu m$)
respective THz spot size and, thus, electric field strength at the tip position for a given position of the recollimation mirror. It should be noted that the THz emission and propagation characteristics will depend on the local spot size and wavefront curvature of the NIR pump beam as well as on the THz frequency. Consequently, the intermediate THz focus (or the virtual THz focus in case of a divergent NIR wavefront) does not necessarily overlap with the NIR focus and will depend on the THz frequency. In particular, the THz spectrum at the tip position thus varies with the STE and recollimation mirror positions. The corresponding THz spectra are shown in Figure S4 in the Supporting Information. However, a detailed discussion of the THz emission characteristics under such conditions is beyond the scope of this work, and further investigations are currently in progress.

**THz amplitude scaling.** An important measure for the applicability of the STE for THz-STM is the achievable THz voltage in the STM. Figure 4a) shows the peak THz voltage versus NIR pump pulse energy for different STE positions at 1 MHz repetition rate. We find that peak THz voltages up to 2 Volts can be achieved when placing the STE 4-5 mm in front of the NIR focus. At closer distances, the incident THz field strength and, thus, the THz voltage decrease due to saturation caused by thermal heating and hence a reduced STE magnetization, as well as due to a decreased collection efficiency of the highly divergent THz beam emitted from sub-wavelength-sized spots. Moreover, ablation and white light generation in the sapphire substrate prohibit the use of higher pump pulse energies at distances very close to the NIR focus in the current setup. This effect can likely be mitigated by modification of the STE excitation geometry with optimized pump spot sizes, i.e. using a weakly focused or collimated NIR pump beam. In addition, plane wave excitation will further allow for more well-defined waveform control via the Gouy phase at constant THz
bandwidth. At distances far away from the NIR focus, the expected linear fluence dependence dominates the signal, and the THz voltage decreases again with larger STE distances due to the increasing NIR pump spot size, as can be seen from Figure 4b). These results demonstrate that high THz voltages can be achieved in the STM using the STE as THz source, but point out the importance of an optimized STE excitation geometry at µJ-level pulse energies and intermediate repetition rates as required for THz-STM.

**Tip-sample distance dependence.** We finally analyze the dependence of the tip-enhanced THz waveform on the tip-sample distance. Figure 5a) shows the dependence of the current...
through the STM junction versus tip-sample distance $d_{\text{rel}}$ relative to the position of a setpoint of 200 pA current and 10 V DC bias. Upon retraction, the current drops rapidly within the first 1 nm, indicating that we are in the tunneling regime with a current composed of DC and photo-assisted tunneling. The onset of tunneling is also evident from the I-V curve shown in Figure 5e) recorded at the setpoint position with the feedback switched off. Note that with these settings, the actual distance to the sample is still several Ångstrom up to a few nanometers, as also indicated by the current recorded in the forward direction (negative $d_{\text{rel}}$). With increasing distances, the photocurrent stays nearly constant until at around 600 nm a pronounced photocurrent peak is observed presumably due to interference effects$^{38}$ of the exciting NIR laser pulse in the junction. Whereas at far gap distances multiphoton photoemission above the broad barrier dominates, see Figure 1b), tunneling of photoexcited electrons through the narrowed barrier at lower energies will contribute and eventually dominate the photocurrent at close gap distances. Disentangling the different contributions requires a detailed analysis of the photocurrent nonlinearity and the potential barrier in the STM junction$^{39}$, which is beyond the scope of this work. Considering the high DC bias of 10 V, it is reasonable to assume that for relative distances larger than 1 nm the photocurrent is dominated by short-lived high-energy electrons and, thus, instantaneous on the time scale of the THz pulse. This assumption is supported by the results discussed below.

THz near-field waveforms and their respective I-V curves are recorded at different distances by retracting the sample a defined step and plotted in Figures 5d) and 5e), respectively. To avoid influence from drift especially at the setpoint distance, the feedback is temporarily switched on again between each THz-NIR time delay to reference the tip position. We find
that the THz waveform does not change considerably over a wide range of tip-sample distances. We further observe a nearly constant THz voltage applied to the STM junction as plotted in Figure 5b). As discussed below, this behavior is expected from the quasi-static nature of the THz field. To better understand the scaling with tip-sample distance, we perform frequency-domain simulations of the tip-enhanced THz field in the junction (using the RF-module of COMSOL Multiphysics, details are described in the Supporting Information). The THz-induced potential difference $U_{\text{THz}}$ applied between tip and sample can then be found by line integration of the tip-enhanced THz field $E_{\text{THz}}$ across the junction,

$$U_{\text{THz}}(\omega, d) = \int_{\text{tip}}^{\text{sample}} E_{\text{THz}}(\omega, d) \cdot dr.$$  \hspace{1cm} (2)

Figure 5c) shows the distance scaling of the normalized THz-induced potential difference between tip and sample (solid blue curves, left y-axis) and peak THz electric field (dashed blue lines, right y-axis) as obtained from simulations for three THz frequencies. The THz electric field at the tip and, thus, the THz field enhancement, strongly increase at shorter distances and closely follow the scaling of the DC electric field obtained from a constant potential difference applied to the STM junction (red solid curve). Consequently, the THz voltage stays approximately constant over the entire distance range. At 1 THz, the calculated potential difference changes by less than 1% over the entire range, whereas at 10 THz the THz voltage reduces by about 8% from 1000 nm to 1 nm. These deviations from a constant potential at higher THz frequencies might indicate the limitations of the quasi-static approach. Within our experimental error, our results, thus, support the quasi-static nature of THz voltage pulses applied to nanoscale junctions, although limitations at high THz frequencies need to be further investigated. It is worth noting that the THz field enhancement strongly
decreases with higher frequencies as discussed in the Supporting Information, in line with the observed low-pass filtering of the THz field by the STM tip. We further emphasize that at very close gap distances the THz waveform might be low-pass filtered by the increased lifetime of photoexcited electrons tunneling at lower energies through the junction, which is not significant at the conditions used here. Given that the undistorted waveform is precisely known, such carrier-induced waveform distortions will in turn allow to study few-femtosecond photocarrier dynamics with far sub-cycle temporal resolution and a spatial resolution given by the localization of the photoexcited current.

In summary, we demonstrated efficient coupling of ultrabroadband single-cycle THz pulses from a spintronic emitter (1-30 THz) to the junction of a scanning tunneling microscope. The STE is an attractive source for THz-STM operation, not only due its high bandwidth and fast field transients, but also due to its simple and potentially fast (tens of kHz) control of the THz polarity and polarization via the STE magnetization, as well as the possibility of phase control via the STE excitation geometry. The ability for tight THz focusing provided by the high bandwidth and beam quality of the STE compensates for its relatively low conversion efficiency, thereby enabling high THz voltages at reduced THz power. We demonstrated that THz voltage transients with frequencies up to 15 THz reaching peak voltages up to 2 Volts can be coupled to the STM junction. Our results further show that the low-pass filtering characteristics of the STM tip antenna is a crucial parameter in the design of ultrabroadband THz-STM when aiming at increased time resolution by applying even faster THz voltage transients. In this regard, we show that broadband phase-resolved detection of the tip-enhanced THz field can be achieved by THz-induced modulation of laser-excited photocurrents through the STM junction. We, thus, demonstrate direct experimental access
to the THz near-field waveform over a broad frequency range, allowing for the broadband characterization of the tip transfer function directly in the STM environment. Moreover, our experimental finding that the THz voltage remains constant over a wide range of tip-sample distances verifies the quasi-static nature of THz pulses coupled to the STM. We believe that even faster voltage transients can be achieved in the STM by resonant enhancement of high THz frequencies via tip antenna design\textsuperscript{14} or by blue-shifting the incident THz spectrum, e.g. via control of the frequency-dependent propagation of the THz beam emitted from the STE. At higher (tens of THz) frequencies we expect retardation effects to influence the enhanced THz near-field. In this regard, our work provides a direct route towards the experimental characterization of the phase and amplitude of multi-THz voltage transients applied to an STM junction with few-femtosecond resolution.
Methods

Experiments are performed in an ultrahigh vacuum (UHV) system (base pressure of $< 5 \times 10^{-10}$ mbar) at room temperature. The STM (customized Unisoku USM-1400 with Nanonis SPM controller) is equipped with two off-axis parabolic mirrors (PM) integrated on the spring-loaded STM platform (1 × bare Au and 1 × protected Ag, 1” diameter, 35 mm focal length). The beams incident angles are 68° with respect to the tip axis. The THz beam enters the UHV chamber via a 500 µm thick diamond window and is focused by the Au mirror. NIR pulses are focused via the Ag PM for photoexcitation of the STM junction. The tip position is fixed and the sample is moved for coarse motion and scanning. The two PMs are motorized and can be moved in xyz-direction (Attocube GmbH) for precise focus adjustment on the tip apex. The DC bias is applied to the sample and the current is collected from the grounded tip. The current amplifier (Femto DLPCA) is operated at a gain of $10^9$ V/A at 1 kHz bandwidth. The THz-induced current is detected by chopping the THz excitation beam incident on the STE at 607 Hz and lock-in detection. The Ag(111) sample was cleaned by repeated cycles of Ar+ sputtering and annealing up to 670 K. Electrochemically etched tungsten tips are transferred to UHV immediately after etching.

A broadband OPCPA laser system (Venteon OPCPA, Laser Quantum) delivering 8 fs VIS-NIR laser pulses (800 nm center wavelength) with 3 µJ energy at 1 MHz repetition rate is used for THz generation and photoexcitation of the STM (2 µJ are available for the THz-STM setup). Part of the laser output is focused by a PM (focal length = 50 mm) for THz generation from the STE (5.8 nm thick W/CoFeB/Pt trilayer on 500 µm sapphire substrate$^{17}$) at normal incidence. The emitted THz radiation is collected by a second PM (focal length =
50 mm) and a 500 µm thick silicon wafer is used to block the collinear NIR pump beam. The THz beam is collinearly overlapped with 8 fs NIR pulses at a variable time delay for electro-optic sampling as well as for precise beam alignment in the STM. A 300 µm thick ZnTe(110) crystal is used for electro-optic detection of the THz field outside UHV in a reference beam path that is identical to the STM beam path. The EOS signal is deconvoluted with the detector response in the time domain\textsuperscript{17,36} to obtain the original THz electric field (EOS signals and deconvoluted fields are shown in the Supporting Information).

Numerical simulations are performed to calculate the THz near-field in the tip-sample junction by solving the time-harmonic wave equation for the electric field within the RF-Module of COMSOL Multiphysics 5.5. Details are provided in the Supporting Information.

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ASSOCIATED CONTENT

Supporting Information. Discussion of space charge effects, dependence of THz waveforms on DC bias and THz voltage, antenna model, measurement of incident THz electric fields, dependence of THz spectra on STE position, numerical simulation of THz electromagnetic field distributions

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Notes

The authors declare no competing financial interest.
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Supporting Information:

Phase-resolved Detection and Control of Ultrabroadband THz Pulses coupled to a Scanning Tunneling Microscope Junction

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1. Discussion of space charge effects

Sampling the THz voltage across the STM junction via THz-induced modulation of the photocurrent requires that the photocurrent reacts quasi-instantaneous on the time scale of the applied THz field. However, several effects such as photoelectron propagation in the tip-sample gap or long-lived hot carriers in the photoexcited tip or sample may disturb the measurement and have to be ruled out. As discussed by Yoshida et al.,⁸ THz-Streaking back into the tip can be circumvented by operating at high DC bias and low THz fields⁸, whereas lifetime effects from hot carriers can be neglected when operating in the regime of multiphoton photoemission above the potential barrier. Another effect, not considered by Yoshida et al.,⁸ is space charge, i.e., the Coulomb repulsion experienced by electrons in a photoelectron cloud containing multiple electrons. Due to the nanometer-sized volume at the tip apex, even a low number of electrons can lead to high photoelectron densities, leading to strong Coulomb interaction between the photoelectrons. As a consequence, ‘leading’ photoelectrons at the front of the space charge cloud are accelerated away from the tip faster than by the DC acceleration alone, whereas ‘backside’ photoelectrons close to the tip surface are accelerated back towards the tip by the space charge cloud in front of them. Those back-accelerated electrons can then be steered back into the tip by THz fields much lower than the

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⁸It should be noted that, even at low THz and high DC fields, the THz field always affects the photoelectron trajectories in the tip-sample gap, depending on the emission time and flight time of the photoelectrons with respect to the arrival and duration of the THz pulse. As we detect the total current and average over all energies and arrival times of the photoelectrons, our measurement is not sensitive to small modulations of the electron trajectories, but is only sensitive to a reduction of the total electron yield by THz-streaking back into the tip.
DC field due to the described effect of space charge acceleration. Moreover, these photoelectrons are not rapidly accelerated away from the tip as expected from the large inhomogeneous DC field at the tip, but can remain a significant amount of time close to the tip surface. Hence, they not only experience the instantaneous THz field at the time of photoemission, but can be steered back into the tip by a THz field at a much later delay within the THz pulse, i.e., the overall process is not instantaneous and can act as an effective low pass filter distorting the measured THz waveform.

In Figure S1 we plot THz waveforms measured in the STM junction at 1 µm gap distance and 8 V DC bias for different photocurrents. The applied THz voltage is 0.12 V at the maximum peak. As can be seen from Figure S1a), the measured waveform clearly depends on the number of electrons excited per laser pulse, with a most pronounced deformation at

![Figure S1](image-url)
the beginning of the pulse before the main half cycle. This is accompanied by a reduction of the THz bandwidth as plotted in Figure S1b), revealing the effective low pass filtering due to space charge. Moreover, Figure S1c) shows that in this regime the shape of the THz waveform also depends on the THz polarity. These deformations and deviations with polarity are most pronounced at the beginning of the THz pulse. This is not surprising as those photoelectrons excited shortly before the arrival of the main pulse experience the strongest THz field, whereas the photoelectrons excited at later times only experience the ringing oscillations after the main THz cycle. For even higher electron numbers, severe waveform distortions are observed as plotted in Figure S1d), with distortions and polarity dependence occurring over the full THz waveform.

Understanding the exact waveform deformations in the presence of space charge requires a detailed analysis of the dynamics of the photoelectron cloud in the tip-sample gap and is beyond the scope of this work. At this point, we conclude that elimination of space charge contributions is crucial for reliable THz waveform sampling by the employed technique as described in the main manuscript. In our experiments we proof the absence of space charge effects to our measurements by systematically reducing the number of photoelectrons to a value at which no waveform distortions and no spectral filtering is observed by further lowering the photocurrent. These observations point out the importance for a careful consideration of non-instantaneous effects potentially disturbing the sampled THz voltage transient.

2. Dependence of THz waveforms on DC bias and THz voltage

The propagation of photoelectrons in the tip-sample gap (and potentially back to the tip) depends on the combined forces of the DC field, the THz field, and space charge interactions. Moreover, waveform distortions from photo-assisted tunneling of electrons close to the Fermi level with longer lifetimes may be expected to contribute at high DC bias and short gap distances. As the lifetime of hot electrons depends on the energy window sampled by the THz pulse, distorting effects from hot carriers should also depend on the DC bias and THz voltage amplitude. Hence, undistorted instantaneous THz near-field sampling requires that the measured THz waveform does not depend on the DC bias and incident THz field strength.
within the linear range of the I-V slope (see Figure 1c)). Figure S2b) shows THz waveforms measured for three different DC biases with the corresponding I-V curve plotted in Figure S2a). All waveforms exhibit the same shape and voltage amplitude as expected from the linear slope of the I-V curve. Figure S2c) shows THz waveforms measured for two incident THz amplitudes at 10 V bias, and again we find that the sampled THz waveform does not depend on the applied THz voltage. In addition to the constant THz waveform observed for opposite THz polarities, as plotted in Figure 1d) in the main manuscript, these results confirm the interpretation and validity of our sampling approach.

3. Antenna model

The model used to fit the tip antenna response in Figure 2 treats the tip as an RLC electronic circuit with resistance $R$, capacitance $C$ and inductance $L$ connected in series. In this very simple model, the incident THz electric field applies a voltage to the antenna that leads to a current induced inside the antenna of

$$I_{\text{THz}}(\omega) \propto \frac{E_{\text{in}}(\omega)}{R + i\omega L - i(\omega C)^{-1}}.$$ 

The 0.25 THz resonance frequency obtained from the best fit values of $R = 102$ $\Omega$, $L = 0.96$ pH and $C = 4.46$ fF is in reasonable agreement with previous results by other groups. The simple RLC model considers only the tip wire alone as the antenna. A more
detailed theoretical analysis of the receiving properties of an STM junction would require a more advanced model taking into account the sample and junction properties in addition to the tip.

4. Measurement of incident THz electric fields
To analyze the THz electric field incident to the STM junction, we pick the THz beam before entering the STM chamber and focus the THz pulses in a 300 µm thick ZnTe(110) crystal for electro-optic sampling (EOS). The path length and optical components are identical to the STM beam path. In EOS, the measured time-domain signal $S(t)$ is given by the convolution of the THz electric field $E_{\text{in}}(t)$ incident on the detector with the detector response function $h_{\text{det}}$,

$$S(t) = (h_{\text{det}} * E_{\text{in}})(t).$$

Hence, if $h_{\text{det}}(t)$ is known, the THz electric field can be obtained by deconvolution of the EOS signal with the detector response, which depends on the properties and thickness of the electro-optic medium and the sampling pulses. After calculation of $h_{\text{det}}(t)$, the deconvolution is performed numerically as described in more detail in references 17,36. Figure S3 shows the deconvoluted waveforms of the THz electric field incident on the STM tip for all four STE positions plotted in Figure 3b) in the main manuscript.

![Figure S3](image)

**Figure S3.** (a) THz electric field transients incident onto the ZnTe(110) detection crystal for four different STE positions as obtained from deconvolution of the EOS signal with the detector response. The phase shift applied by moving the STE inside the focused NIR beam is apparent in the waveforms. (b) Corresponding EOS signals (bottom) and detector response function $h_{\text{det}}(t)$ (top).
Direct comparison of the THz electric field obtained from EOS and the THz voltage waveform measured in the STM requires that both waveforms are recorded at the same position along the focused THz beam. In the EOS setup, the detection crystal is positioned in the focus of the tightly focused NIR beam that propagates collinear with the THz beam and is used as sampling pulse. Likewise, we use the same collinearly propagating NIR pulses as alignment beam to adjust the THz focus on the tip in the STM. Precise alignment is hereby ensured by monitoring the light reflected off the Ag sample and collected via the second (Ag) parabolic mirror inside the STM chamber, and by optimization of NIR-induced photoemission from the tip apex.

5. Dependence of THz spectra on STE position

As discussed in the main manuscript, the THz spectra can vary with STE position due to a frequency-dependent generation and collection efficiency of the THz radiation when generated with curved NIR wavefronts. Figures S4a) and S4b) show the THz amplitude spectra of the tip-enhanced THz waveforms plotted in Figures 3b) and 3c), respectively. We observe a slight bandwidth reduction upon moving the STE away from the NIR focus, as well as when moving the recollimation mirror away from its reference position (defined by recollimation of the NIR pump beam). Such variation of the THz spectrum can originate from the frequency-dependent Gouy phase shift experienced by the THz pulse upon propagation.

Figure S4. Fourier amplitude spectra of the tip-enhanced THz waveforms (a) vs STE position shown in Figure 3b), and (b) vs position of the THz recollimation shown in Figure 3c). We observe a red-shift of the spectra and reduced THz bandwidth when moving the STE away from the focus and when moving the recollimation away from its reference position (details see text) due to a frequency-dependent THz propagation.
to the far field when excited by a curved NIR wavefront, and by a frequency-dependent recollimation, propagation and focusing of the THz beam into the STM.

6. Numerical simulation of THz electromagnetic field distributions

Numerical simulations are performed to calculate the THz response of the STM tip by solving the time-harmonic wave equation for the electric field within the RF-Module of COMSOL Multiphysics 5.5. For a given excitation frequency, COMSOL solves for the full time-harmonic electromagnetic field distribution. Static field distributions are calculated using the AC/DC module of COMSOL. Simulations are performed in 3D with the tip oriented along the z-axis. The tip is modelled by a conical wire with a half opening angle of 4° terminating in an apex of 50 nm radius. The sample of 10 µm thickness is placed at a variable distance $d$.

![Figure S5. Spatial distributions of the tip-enhanced THz electric field (normalized field) in the tip-sample junction at 1 nm and 100 nm gap distances for frequencies of (a-b) 1 THz and (c-d) 10 THz, respectively. The color bars represent the enhancement of the THz field inside the junction with respect to the incident THz field amplitude. The red arrows indicate the direction and polarization of the incident THz field. Simulation parameters are described in the text.](image-url)
in front of the tip. The width of the simulation volume is 200×200 µm and the wire is truncated by the end of the simulation volume at a height of 200 µm from the apex. The simulation volume is surrounded by perfectly matched layers to absorb all outgoing waves. The tip-sample junction is illuminated by plane-wave THz radiation propagation along the $x$-$z$-direction at an angle of 68° with respect to the tip axis as and is polarized linearly in the $x$-$z$-plane, as indicated by the red arrows in Figure S5b). The simulation volume is cut in half along the $y$-direction (out of plane) according to the symmetry given by the THz beam direction to reduce the computational cost. The materials properties of the tip and sample are determined from the complex dielectric functions of tungsten and silver, whose real and imaginary parts are calculated from the Lorentz-Drude model with the Lorentz-Drude parameters taken from Rakic et al.\textsuperscript{40}.

Figures S5a)-d) show spatial distributions of the tip-enhanced THz electric field in the tip-sample junction for two THz frequencies. As can be seen from the color scales, the THz field enhancement increases significantly with lower THz frequencies and shorter gap distances. At far distances b) and d), the strongly inhomogeneous THz near-field is confined to the tip apex with a spatial extent given by the tip radius, and decays rapidly with larger distances away from the tip surface. At nanometer distances much smaller than the tip radius, the field becomes spatially more localized inside the tip-sample gap, and becomes homogeneous like in a plate-capacitor at the junction center $(x,y) = (0,0)$. We obtain the THz-field-induced potential difference from the computed fields by line integration of the $z$-component of the THz near-field along the center of the junction at $(x,y) = (0,0)$. We confirmed that line integration along other pathways yields the same THz-induced potential difference.
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