Terahertz STM for Imaging Ultrafast Nanoscale Dynamics

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Abstract: We couple terahertz (THz) pulses to the tip of a scanning tunneling microscope (STM) and observe THz-pulse-induced tunneling in an STM, enabling a new ultrafast technique called THz-STM that allows for direct imaging of sub-picosecond dynamics on surfaces with nanometer spatial resolution. Imaging of sub-picosecond carrier capture dynamics into a single semiconductor nanodot is demonstrated. The potential of THz-STM for imaging ultrafast phenomena with atomic resolution is discussed.

OCIS codes: (320.7160) Ultrafast technology; (300.6495) Spectroscopy, terahertz; (320.7130) Ultrafast processes in condensed matter, including semiconductors.

1. Introduction

Experimental tools that can spatially resolve ultrafast phenomena over nanometer length scales are essential for understanding nanoscale excitation dynamics in materials as well as for measuring switching speeds of nanodevices. The scanning tunneling microscope (STM) uses quantum tunneling of electrons between a scanning tip and a sample to achieve nanoscale imaging of surfaces with atomic resolution [1]. However, the time resolution of an STM is typically limited by the bandwidth of the amplifier electronics used to measure the small tunnel currents. This has spurred the development of various ultrafast STM techniques that combine STMs with femtosecond laser sources. Photoconductive gating of specially designed GaAs scanning probe tips has achieved 2 ps time resolution and 20 nm spatial resolution [2], but the spatial resolution in this approach is limited by capacitive coupling effects [3]. Junction-mixing STM uses femtosecond lasers to photoconductively generate picosecond voltage pulses along microstrip transmission lines patterned onto the sample. The nonlinear STM tunnel current with bias results in a voltage-pulse-induced tunnel current, achieving 1 nm spatial resolution and a time resolution on the order of 10 ps limited by the bandwidth of the microstrip transmission line [4,5]. Directly focusing femtosecond laser pulses onto the tunnel junction can modify the tunnel current via sample excitation but can also result in thermal expansion of the STM tip [6,7], unless special pulse delay schemes are employed [8]. Recent advances in scattering scanning near-field optical microscopy (s-SNOM) using mid-infrared probe pulses scattered from an AFM-like tip have achieved 20 nm spatial resolution and sub-ps time resolution of ultrafast plasmon dynamics in graphene [9].

2. THz-STM

Femtosecond laser sources have been used extensively to generate picosecond, single-cycle terahertz (THz) pulses for probing materials [10,11]. In a new approach to ultrafast STM called THz-STM, as shown schematically in Fig. 1(a), we have coupled THz pulses generated by an ultrafast laser source (Coherent RegA, 800 nm, 70 fs, 250 kHz) to the tip of a commercial STM (RHK UHV SPM 3000) and observed for the first time THz-pulse-induced tunneling in an STM [12]. A THz pulse coupled to the STM tip acts like a fast voltage transient across the tunnel junction that results in a rectified tunnel current signal, similar to junction-mixing STM. THz pulse autocorrelations performed on gold-on-graphite samples show THz-STM response times of less than 400 fs with simultaneous 2 nm spatial resolution under ambient conditions. We used the Simmons model for the STM tunnel current as a function of bias [13] to describe the observed THz-STM signals. The THz-STM accesses an ultrafast tunneling field emission regime that we are exploring further. In these experiments, we made no modifications to either the sample or STM tip. Efforts are currently underway to get the THz-STM operational in UHV in order to achieve atomic resolution.

3. Imaging ultrafast carrier capture in a single InAs nanodot

Optical-pump/THz-STM-probe experiments were performed on InAs nanodots deposited by MBE on GaAs. The typical size of the nanodots was around 100 nm x 50 nm. 800 nm, 150 fs pump pulses were focused onto the sample region under the STM tip, and THz probe pulses coupled to the tip were modulated by a chopper. The experiment is described in Fig. 1, where electron capture into the InAs nanodot at positive time delays changes the THz-STM signal. Figure 2 shows the carrier capture dynamics over a single InAs nanodot.

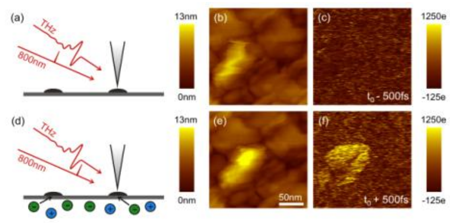

Fig. 1. Using THz-STM to probe ultrafast carrier capture dynamics in a single InAs nanodot on GaAs. (a) THz pulse arrives before an 800 nm, 150 fs pump pulse (negative pump-probe delay time). (b) STM topography scan of the InAs nanodot taken simultaneously with the THz-STM signal (c) at a delay time of -500 fs. The THz-pulse-induced signal is almost zero over both the InAs nanodot and GaAs substrate. (d) The pump pulse excites photocarriers in the GaAs substrate. Electrons are captured quickly, followed by the capture of holes a few picoseconds later. The THz pulse arrives afterwards (positive pump-probe delay time). The InAs nanodot (e) at early times (+500 fs) after photoexcitation gives a THz-STM signal (f) over the nanodot. The scale for the THz-STM signal in (c) and (f) is given as the number of rectified tunnel electrons induced by each THz pulse. The voltage bias was -3.5 V, the current setpoint was 0.5 nA, and the peak THz field was 0.10 kV/cm for all images. [12]

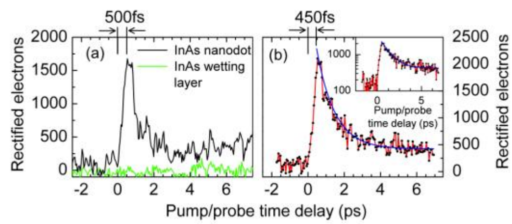

Fig. 2. Optical-pump/THz-STM-probe time scans of ultrafast carrier capture dynamics of single InAs nanodots. (a) Pump-probe time scan with a rise time of the pump-induced signal of about 500 fs. (b) Pump-probe time scan over a different InAs nanodot with a rise time of 450 fs. The blue line is a fit to the data by an exponential decay with a constant offset. The inset shows the same pump-probe response on a semi-log plot. [12]

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5. References

[1] C. J. Chen, *Introduction to Scanning Tunneling Microscopy* (Oxford Univ. Press, 1993).

[2] D. A. Yarotski et al., "Ultrafast carrier-relaxation dynamics in self-assembled InAs/GaAs quantum dots," J. Opt. Soc. Am. B **19**, 1480 (2002).

[3] R. H. M. Groeneveld and H. van Kempen, "The capacitive origin of the picosecond electrical transients detected by a photoconductively gated scanning tunneling microscope," Appl. Phys. Lett. **69**, 2294-2296 (1996).

[4] G. M. Steeves, A. Y. Elezzabi, and M. R. Freeman, "Nanometer-scale imaging with an ultrafast scanning tunneling microscope," Appl. Phys. Lett. **72**, 504-506 (1998).

[5] N. N. Khusnatdinov, T. J. Nagle, and G. Nunes, Jr, "Ultrafast scanning tunneling microscopy with 1 nm resolution," Appl. Phys. Lett. **69**, 2294-2296 (1996).

[6] S. Grafström, "Photoassisted scanning tunneling microscopy," J. App. Phys. **91**, 171-1753 (2002).

[7] A. Dolocan et al., "Two-color ultrafast photoexcited scanning tunneling microscopy," J. Phys. Chem. C **115**, 10033-10043 (2011).

[8] Y. Terada, S. Yoshida, O. Takeuchi, and H. Shigekawa, "Real-space imaging of transient carrier dynamics by nanoscale pump-probe microscopy," Nature Photon. **4**, 869-874 (2010).

[9] M. Wagner et al., "Ultrafast and nanoscale plasmonic phenomena in exfoliated graphene revealed by infrared pump-probe nanoscopy", Nano Lett. Doi/10.1021/nl4042577 (2014).

[10] P. U. Jepsen, D. G. Cooke, and M. Koch, "Terahertz spectroscopy and imaging – Modern techniques and applications," Laser Photon. Rev. **5**, 124-166 (2011).

[11] R. Ulbricht, E. Hendry, J. Shan, T. F. Heinz, and M. Bonn, "Carrier dynamics in semiconductors studied with time-resolved terahertz spectroscopy," Rev. Mod. Phys. **83**, 543-586 (2011).

[12] T. L. Cocker, V. Jelic, M. Gupta, S. J. Molesky, J. A. J. Burgess, G. De Los Reyes, L. V. Titova, Y. Y. Tsui, M. R. Freeman, and F. A. Hegmann, "An ultrafast terahertz scanning tunnelling microscope," Nature Photon. **7**, 620-625 (2013).

[13] J. G. Simmons, "Generalized formula for the electric tunnel effect between similar electrodes separated by a thin insulating film," J. Appl. Phys. **34**, 1793-1803 (1963).