Imaging ultrafast dynamics on the nanoscale with THz-STM

V. Jelic[1], T. L. Cocker[1§], P. H. Nguyen[1], C. Rathje[2], G. J. Hornig[1], J. R. Hoffman[1], H. M. Sharum[1], R. Miller[1], S. J. Molesky[1], M. Gupta[3], J. A. J. Burgess[1†], G. B. De los Reyes[1], L. V. Titova[1ǂ], C. Ropers[2], Y. Y. Tsui[3], M. R. Freeman[1], and F. A. Hegmann[1]*

[1]Department of Physics, University of Alberta, Edmonton, AB, T6G 2E1 Canada

[2]4th Physical Institute, University of Göttingen, 37077 Göttingen, Germany

[3]Department of Electrical and Computer Engineering, University of Alberta, Edmonton, AB, T6G 2E1 Canada

[§]Current address: Department of Physics, University of Regensburg, 93040 Regensburg, Germany

[†]Current address: Max Planck Institute for the Structure and Dynamics of Matter, 22761 Hamburg, Germany

[ǂ]Current address: Department of Physics, Worcester Polytechnic Institute, Worcester, MA, 01609 USA

*hegmann@ualberta.ca

***Abstract*—The ability to directly image ultrafast phenomena with nanometer spatial resolution is essential to our understanding of local excitation dynamics in nanomaterials and devices. We have developed a new approach to ultrafast scanning tunneling microscopy (STM) that couples terahertz (THz) pulses to the scanning tip of a STM. We have used THz-STM under ambient lab conditions to image ultrafast charging dynamics of a single InAs nanodot on GaAs with 0.5 ps time resolution and 2 nm spatial resolution. We are currently developing THz-STM for operation in ultrahigh vacuum with the goal of imaging ultrafast dynamics on surfaces with atomic resolution.**

I. INTRODUCTION

The scanning tunneling microscope (STM) can image surfaces with atomic resolution, but the time resolution is typically limited to a few tens of microseconds by the bandwidth of the amplifier electronics. To achieve picosecond or sub-picosecond time resolution, various ultrafast STM techniques that integrate femtosecond lasers have been developed [1-8]. Time-resolved terahertz (THz) pulse spectroscopy is now a well-established technique for probing ultrafast carrier dynamics in nanomaterials [9,10], but the spatial resolution is typically set by the diffraction limit spot size of the THz pulse (~ 1 mm). Recently, however, scattering scanning near-field optical microscopy (s-SNOM) using infrared and multi-THz pulses has enabled imaging of ultrafast carrier dynamics in graphene [11] and ultrafast photoconductivity dynamics in single semiconductor nanowires with 10 nm spatial resolution [12].

.

II. ULTRAFAST THZ-STM

As shown schematically in Figs. 1 and 2, a new ultrafast STM technique that couples THz pulses to the scanning probe tip of an STM has been demonstrated (THz-STM), providing simultaneous 0.5 ps time resolution and 2 nm spatial resolution under ambient laboratory conditions [13-15]. The THz pulse acts like a fast voltage transient across the tunnel junction that results in a rectified tunnel current signal. Optical pump – THz-STM probe studies can also be performed, as shown in Fig. 2, as demonstrated by imaging ultrafast carrier capture into a single InAs nanodot on GaAs photoexcited with 800 nm pump pulses [13-15] (Fig. 3).

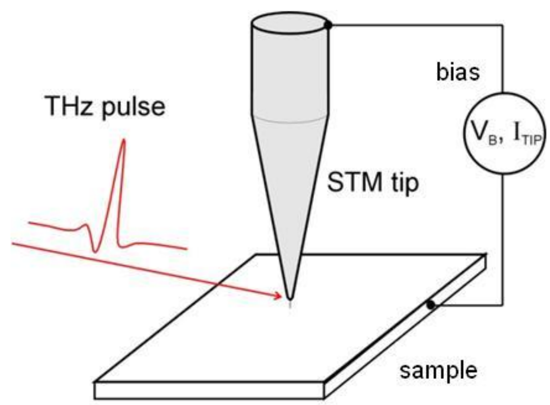

Fig. 1. Schematic of THz-STM showing THz pulse coupling to the scanning tip of an STM.

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. 3, where electron capture into the InAs nanodot at positive time delays changes the THz-STM signal. Figure 4 shows the carrier capture dynamics over a single InAs nanodot.

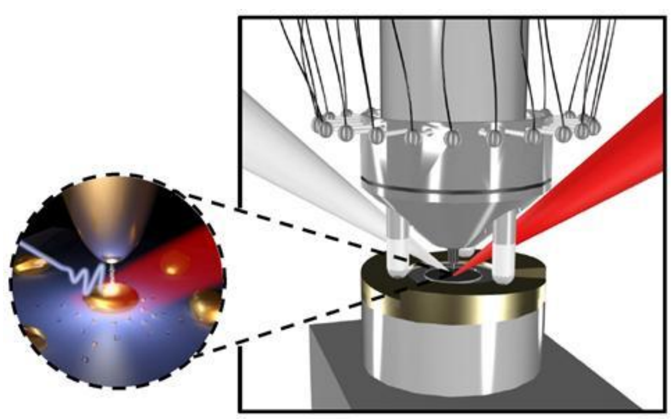

Fig. 2. Schematic of an ultrafast optical pump – THz-STM probe experiment. The STM tripod peizo scan head sits on top of a sample puck.

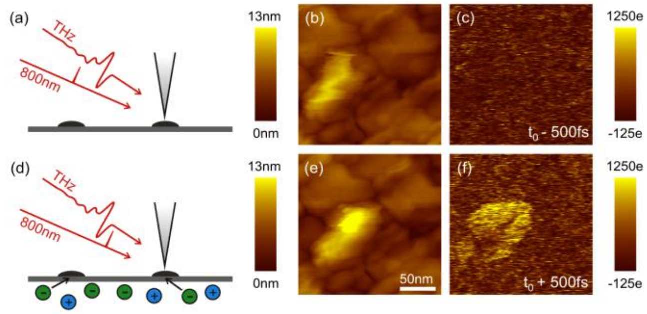

Fig. 3. Probing ultrafast carrier capture dynamics in a single InAs nanodot on GaAs. (a) THz pulse arrives before an 800 nm, 150 fs pump pulse (negative pump-probe delay time). (b) STM topography scan of the InAs nanodot taken simultaneously with the THz-STM signal (c) at a delay time of -500 fs. The 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. [13]

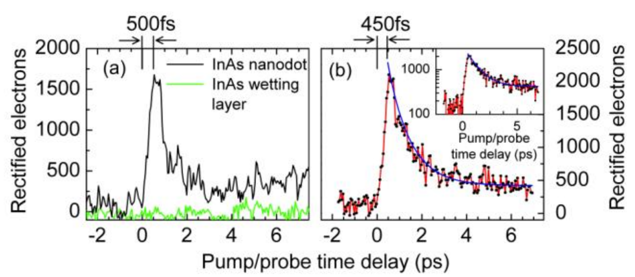

Fig. 4. (a) Optical-pump/THz-STM-probe time scan of ultrafast carrier capture dynamics in a single InAs nanodot. The rise time of the signal is 500 fs. (b) Pump-probe time scan over a different InAs nanodot with a rise time of 450 fs. The blue line is a fit to the data by an exponential decay with a constant offset. The inset shows the same pump-probe response on a semi-log plot. [13]

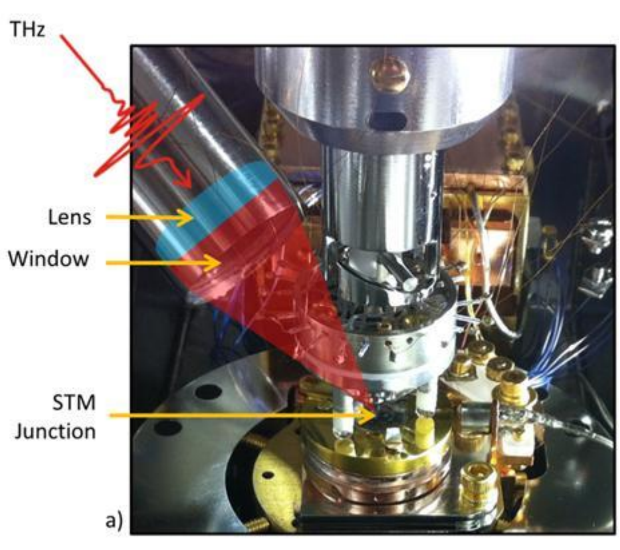

Fig. 5. Photo of the THz-STM system in UHV.

THz-STM is currently being developed for operation in ultrahigh vacuum (UHV) with the goal of achieving imaging of ultrafast nanoscale dynamics with atomic resolution. A TPX lens at the end of an inverted viewport allows focusing of the THz beam close to the STM tip in UHV, as shown in Fig. 5.

III. Acknowledgments

We acknowledge support from NSERC, CFI, ASRIP, iCORE, AITF iCiNano, and the AITF Strategic Chair Program.

.

References

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

[2]. 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).

[3]. 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).

[4]. 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).

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

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

[7]. 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).

[8]. S. Yoshida, Y. Aizawa, Z. Wang, R. Oshima, Y. Mera, E. Matsuyama, H.Oigawa, O. Takeuchi, H. Shigekawa, “Probing ultrafast spin dynamics with optical pump-probe scanning tunneling microscopy,” *Nature Nanotech*. **9**, 588 (2014).

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

[10]. L. V. Titova, T. L. Cocker, D. G. Cooke, X. Wang, A. Meldrum, and F. A. Hegmann, "Ultrafast percolative transport dynamics in silicon nanocrystal films," *Phys. Rev. B* **83**, 085403 (2011)

[11] M. Wagner, Z. Fei, A. S. McCleod, A. S. Rodin, W. Bao, E. G. Iwinski, Z. Zhao, M. Goldflam, M. Liu, G. Dominguez, M. Thiemens, M. M. Fogler, A. H. Castro Neto, C. Ning Lau, S. Amarie, F. Keilman, and D. N. Basov, “Ultrafast and nanoscale plasmonic phenomena in exfoliated grapheme revealed by infrared pump-probe nanoscopy”, *Nano Lett.* **14**, 894 (2014).

[12]. M. Eisele, T. L. Cocker, M. A. Huber, M. Plankl, L. Viti, D. Ercolani, L. Sorba, M. S. Vitiello, and R. Huber, "Ultrafast multi-terahertz nano-spectroscopy with sub-cycle temporal resolution", *Nature Photon*. **8**, 841 (2014).

[13]. 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).

[14]. T. L. Cocker, V. Jelic, J. R. Hoffman, M. Gupta, R. Miller, S. J. Molesky, J. A. J. Burgess, G. De Los Reyes, L. V. Titova, Y. Y. Tsui, M. R. Freeman, and F. A. Hegmann, “Imaging ultrafast dynamics on the nanoscale with a THz-STM”, *Proc. SPIE* **8984**, 898411 (2014).

[15]. V. Jelic, T. L. Cocker, J. R. Hoffman, M. Gupta, R. Miller, S. J. Molesky, J. A. J. Burgess, G. De Los Reyes, L. V. Titova, Y. Y. Tsui, M. R. Freeman, and F. A. Hegmann, “Ultrafast THz-pulse-induced tunneling dynamics in an STM,” *Proc. SPIE* **8984**, 898412 (2014).