WHAT TO DO WHEN IT'S TOO FAST TO SEE: FREEZE-FRAME IMAGING, FROM RACEHORSES TO ATOMS

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ABSTRACT

This paper is an introduction to the field of "ultrafast" scanned probe microscopy. In this field the dream is to combine methods from time-resolved spectroscopy and scanning probe microscopy in order to study the dynamics of physical systems right down to the atomic level. Will it be possible to produce "movies" starring individual atoms?

During the past two decades, time-resolved laser spectroscopy has evolved to the point where phenomena on time scales as short as ten femtoseconds (10^{-14} s) can be observed in the laboratory. Such astounding temporal resolution is sufficient even to study chemical reactions in progress, at the level of "watching" the molecules dissociate. In such experiments there is no spatial resolution to speak of, however. The measurements are performed on macroscopic quantities of molecules and the laser spot, even if it is brought to a diffraction-limited optical focus, is orders of magnitude larger than an individual molecule.

Fortunately, it seems possible to adapt ultrafast laser methods to most of the scanned probe microscopies described elsewhere in this issue, yielding tools with fine temporal *and* spatial resolution. An overview is presented, followed by more discussion of the particular case of ultrafast scanning tunneling microscopy.

"The great panorama of life is interesting because it moves."[1]

INTRODUCTION AND HISTORICAL CONTEXT

There is considerable excitement today in an area of research in the physical sciences where the efforts of "ultrafast" spectroscopists and "ultrafine" microscopists overlap. Each group would like to see the strengths of both combined into a single measurement technique. The 1980's saw time-resolved spectroscopy evolve to the point where chemical reactions such as the photodissociation of simple molecules could actually be experimentally observed in progress as they occurred on femtosecond time scales.[2,3] Similarly, the field of scanning microscopy exploded in the 80's with the introduction of methods for mapping out the equilibrium positions of atoms on surfaces.[4,5] Scanning probe microscopes measure quantities such as the tunneling current between a surface and a probe atom brought into very close proximity with the surface, or even the van der Waals force between the probe atom and the surface. It is now the dream of numerous researchers to combine such methods and study the dynamics of physical systems right down to the atomic level, through observations which amount to "motion pictures" of the action.

When this goal is finally achieved, we will have reached a major milestone in an adventure that began well over 100 years ago, when interest in fast motion (faster than the eye can follow) helped stimulate the development of "instantaneous" photography. Although the associated technologies have changed dramatically since then, permitting studies to proceed to ever-shorter time and length scales, the spirit of the investigations has remained constant.

The ultimate goal of work on ultrafast scanned probe microscopy is the direct observation of dynamics at atomic length scales. Although we are by now accustomed to "still" images with atomic spatial resolution, the prospect of atomic "movies" continues to be met with incredulity. It is of interest therefore to be reminded that photography was at a similar juncture around 1850. At that time, about one minute was a typical time required for a photographic exposure in ambient light. The concept of "flash" was not widely known, and hence it had not become recognized that it might be possible to actually photographically record *phenomena that took place too rapidly for the unaided human eye to distinguish!*

FAST IMAGING

a) Flash - "Split second" viewing well pre-dates the first technology to capture such images, i.e. photography. It is recorded that Sir Charles Wheatstone pointed out in 1834 that the speed of electric light could be used to freeze fast motion, but who is to say what variety of scenes have been accidentally observed for millenia thanks to lightning

bolts through the dark? However, in a proof-of-principle demonstration in 1851, twelve years after the invention of photography, William Henry Fox Talbot used a spark discharge from a bank of Leyden jars to capture an astonishing, readable image of text from a sheet of the London *Times* which had been attached to a rapidly spinning wheel.[6] Unfortunately this achievement was to have little impact on the future development of fast imaging, but Fox Talbot understood the implications and declared, "It is in our power to obtain pictures of all moving objects, no matter how rapid their motion may be, provided we have the means of sufficiently illuminating them with a sudden electric spark."[7] After almost 150 years, this statement from the earliest pioneer of high-speed photography remains an accurate summation. What Fox Talbot could not have imagined is how developments in optics would lead to pulses of light so quick that they give us pause to reconsider what we mean by a pulse of light. Soon these pulses may be cut down to less than one optical cycle (less than one complete wavelength in spatial extent!) A very rough interpretation of this progress across the decades is given in Fig.1. In order to qualify for achieving a certain temporal resolution on this chart, it is necessary not only that the "flash" is sufficiently short, but also that the ability to synchronize the flash to an event be comparably precise. The present day extreme is that it now appears to be in our power to take pictures of even the most rapid of moving objects – invariably the smallest, too small to be seen using visible light – using something akin to a "sudden electric spark", applied to a scanning tunneling microscope.

Flash photography received a great deal of attention with the publication of Eadward Muybridge's work, which resolved the question, 'Is there a point in the stride of a racehorse at which all four hooves are off the ground?' (there is.)[8] Muybridge's work was commissioned by Leland Stanford to settle a racetrack wager, in an episode that became famous in the histories of both photography and of the study of animal locomotion. Previous elaborate studies involving wiring up of horse and rider and installation of sensors along the track had proved inconclusive, so the direct imaging approach was a real breakthrough. A cartoon reaction to the Muybridge photos from the 1878 is shown in Fig. 2 (along with a more recent photo as a reminder of how commonplace such images are today with high shutter speeds and fast film.)

The flash duration for Muybridge's exposures (in the 10 ms range, using photochemical powder), while orders of magnitude shorter than the exposure times of typical photographs of the day, were still very long in comparision to those of ordinary consumer camera flashes today. Ernst Mach and his son Ludwig in the 1880's and 90's highly refined the imaging of super-

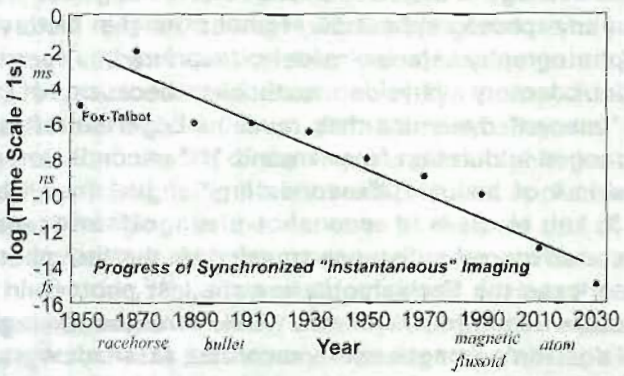

Fig. 1 A highly schematic chart of progress in "instantaneous" imaging across the decades. It's thought that Fox-Talbot had a 10 μsec spark, but it was not synched to the rotation of the wheel. The Machs had μsec flashes. Edgerton reached 10 ns.

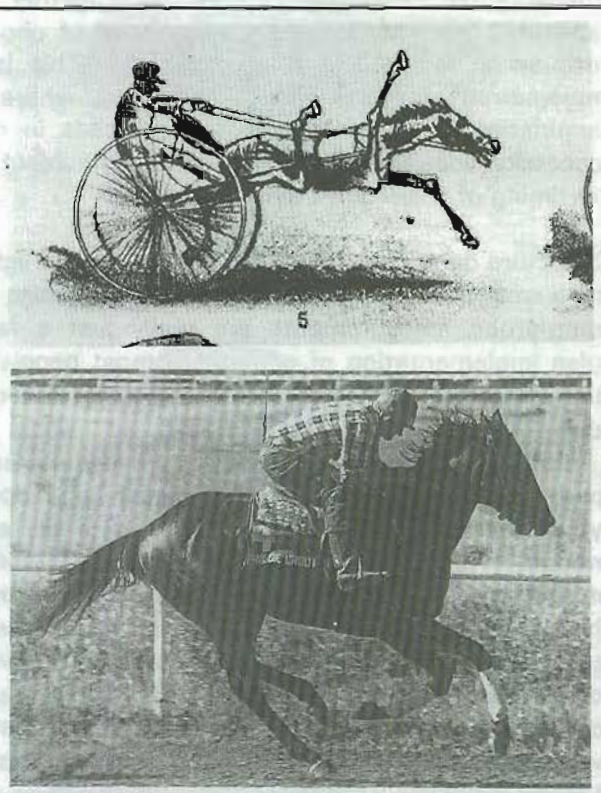

Fig. 2 (top) Part of a cartoon from the *Illustrated Wasp* of 1878 indicating the initial reaction to Muybridge's photos. [from Ref. 1, reprinted with generous permission from the publisher.] (bottom) Typical flash photo we now take for granted , reprinted with permission from the *Edmonton Journal*, Feb. 26 1995, p. E4.

sonic projectiles (bullets and shells) and study of the shock waves using mechanically triggered electric sparks.[9] The familiar modern flash technology was largely developed in the work of Harold Edgerton and colleagues at the Massachusetts Institute of Technology in the 1930's and 40's.[10] Edgerton made many photographs now famous in the history of photography (and widely reprinted, even in introductory physics textbooks because of the "unseen" dynamics they reveal). Edgerton's flashes ranged in duration from around 10^{-5} seconds down to a low of about 10^{-8}seconds (in "length" from about 3 km to 3 m if you shot them off into space, considering the distance traveled by the first photons to leave the flashlamp before the last photons in the pulse emerge.) In the submicrosecond regime Edgerton's images were recorded as shadowgraphs because the total number of photons in the flash was insufficient to expose the film with light reflected by the subject in the standard way.

b) Strobe - To successfully proceed to shorter time scales, it is possible to perform the photographic "measurement" repetitively, adding together the exposures from separate small numbers of photons until an adequate image is obtained. This is an implementation of *stroboscopic* imaging, where the illumination is by a train of short flashes in rapid succession (ideally with the motion of the subject and the timing of the flashes synchronized.)

For lecture demonstrations, an ordinary strobe light[11] and a small electric fan can be used to emphasize that pump-probe measurements are really just a faster pulse implementation of something most people are already well familiar with. Present day mode-locked pulsed lasers produce flashes as short as 5 femtoseconds in duration (about 3 micrometers in spatial extent), corresponding to just a few optical cycles for red light. Such pulses are actually short enough even to record stroboscopic time-domain information about electron dynamics in atomic orbitals. The pulses from the demonstration stroboscope, while terribly long in comparison (20 μs, or 6 km) serve to completely freeze the apparent motion of the fan (a complete blur when otherwise illuminated), making even small scratches on the blades clearly visible. To illustrate dynamics, strings long enough to collide with the bench-top can be taped to opposite blades (for balance), as illustrated in Fig. 3a. The strings have variable tension along their lengths from the rotation, and one can see pulses propagate along the strings from the impacts with the table. Damped oscillations of the ends of the strings are also visible. The dynamics through one complete cycle can be carefully observed by making the strobe frequency slightly slower (or faster) than that of the fan (this is actually most fun when the strobe is flashed a bit faster so the fan appears to move backwards.)

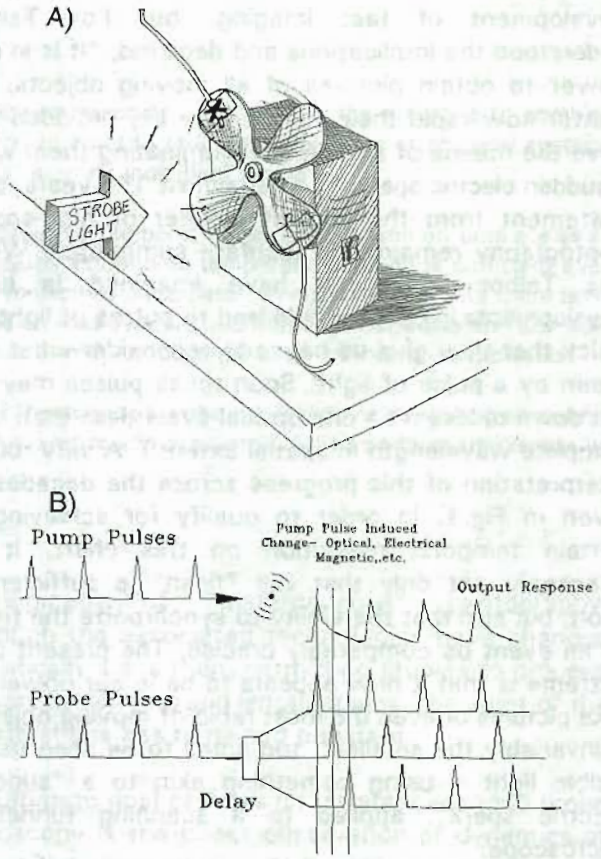

Fig. 3 (a) Sketch of a strobe and fan lecture demonstration to illustrate the idea on a longer time scale (freehand drawing by David Fortin.) (b)Schematic of the arrangement of optical pulse trains in an ultrafast pump-probe measurement

The very short time scales involved in actual ultrafast measurements necessitate slight changes in implementation in comparison to the strobe demo, as indicated in Fig. 3b. The dynamics must be very well synchronized to the optical pulses in order to avoid loss of temporal resolution, and in practice are usually optically "triggered" (directly or indirectly). The timing of the probe light relative to that of the dynamics is changed through optical path lengths, usually controlled using "optical delay lines" consisting of retroreflectors mounted on linear translation stages. But the principle is basically the same.

ULTRAFAST MICROSCOPY

Pulse laser techniques were first and are still most often used to measure various excitation lifetimes in uniform samples, requiring little or no imaging beyond focussing the light when the specimens are smaller than the beam. Of course, time-resolved optical microscopy can also be performed. There are two main implementations, full-field and scanned imaging. In the former, the ordinary view through a microscope is captured using film or a CCD camera. In scanned imaging, the light is brought to a sharp focus and raster scanned over an area of the sample. That is, an image is generated from measurements taken at each position on a rectangular grid. In practice, the sample is often scanned under the focus spot.

Full-field imaging offers the possibility of recording an image using a single pulse of light, a big advantage for studying phenomena having a random component which does not reproduce exactly each time the measurement cycle is initiated. However, it is often necessary to average the signal from multiple pulses to improve the signal-to-noise ratio, which will also smear out spatial details of any random response from the image. If a time-resolved "movie" is to be made from a series of "single-shot" images, the random component will not repeat properly from frame to frame, but at least it will not be overlooked! For full-field imaging, it cannot be doubted that even the shortest optical pulses will eventually be amplified to sufficient intensities to capture single-shot images (although the peak power in the pulses will exceed the damage threshold of some samples.)

Scanned imaging offers a slightly different set of strengths. Weak signal detection is more easily optimized using photomultiplier- or photodiode-based setups. Full time-dependent information is often necessary only at selected spots on a sample after a few images have been taken, in which case data acquistion is much more efficient. Most significantly, perhaps, "near-field" optical imaging for beating the diffraction limit of resolution[12] must be scanned. Illumination through small (sub-wavelength) apertures and scattering from small particles (or the end of a sharp tip) are the presently favoured means of improving resolution to one-tenth (aperture) or possibly even one-one-hundredth (scattering) of the wavelength. One of the biggest challenges is to improve on the tradeoff between spatial resolution and signal strength. More modest "superresolution" imaging (to perhaps $\lambda/4$, and with good optical efficiency) is possible scanned or full-field with the solid immersion lens.[13]

The power of optics is vast in that many different imaging mechanisms are also possible. Beyond the spectrum and intensity of reflected or transmitted light (as recorded by our eyes or on film), the polarization state can be analyzed to reveal a great deal of information about the sample. In Fig. 4 we show time-resolved *magnetic* images of samples from scanning Kerr effect (polarization rotation) measurements. In Fig 4a, a picosecond snapshot of the perpendicular magnetization in an 8 um diameter permalloy disk is shown, from studies of the ferromagnetic dynamics of small particles.[14] Fig 4b shows a practical application of such imaging, to the response of the pole tips of a tiny horseshoe electromagnet used to record data on a hard drive.[15] The speed of such devices must improve along with all the other system components in order for present performance trends to continue, and time-resolved imaging is a direct way to characterize these components. We have made what is perhaps

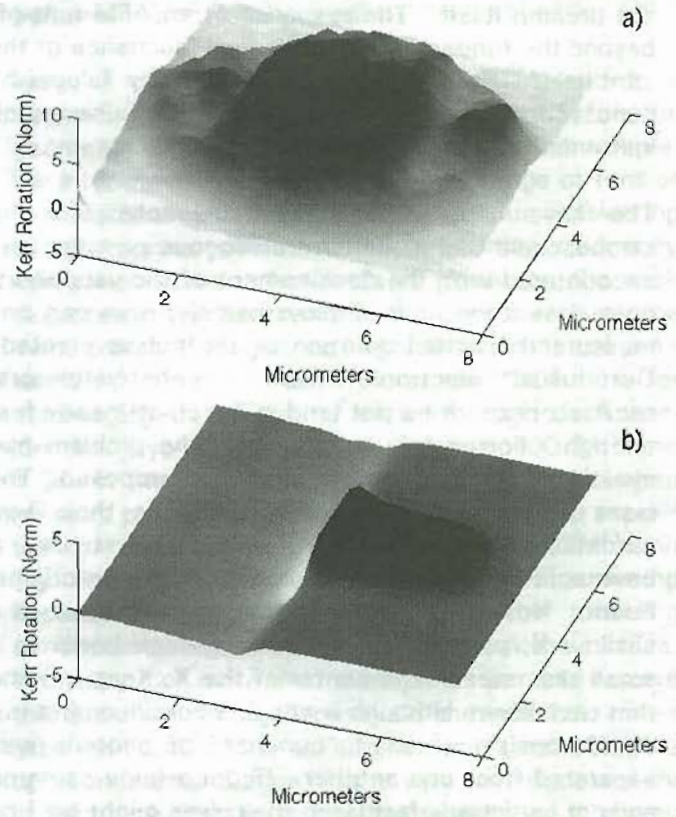

Fig. 4 Time-resolved magnetic imaging. These images are of the magnetization component perpendicular to the surface, showing (a) nonuniform spatial structure observed during measurements of magnetic resonance in a ferromagnetic disk of 8 μm diameter, 100 nm thick permalloy; (b) the magnetization at the (asymmetric) pole tips of a permalloy recording head showing an instant during a transition corresponding to the writing of one bit of data.

the first "picosecond movie", showing two bits of data coming through such a recording head at an effective frame rate of 20 billion frames/sec.[16]

For the "ultimate" in spatial resolution (down to the atomic level) non-optical microscopes are required. There are many possibilities, including picosecond pulsed scanning electron microscopy (some very nice work was performed in the 1980s, for example[17]). We can also anticipate very nice developments in x-ray imaging combined with compact sources of pulsed x-rays. For the remainder of this paper, however, we will emphasize scanning tunneling microscopy.

ULTRAFAST SCANNING PROBE MICROSCOPY

a) Principle - The family of scanning tunneling and atomic force microscopes generally measure signals somewhere in the 1 to 1000 kHz range. In an STM, faster signals are lost to the capacitance of the cable connecting the tip to the preamp, or to the speed of the preamp itself. The response of an AFM rolls-off beyond the fundamental mechanical resonance of the cantilever. How then are even relatively "sluggish" nanosecond-scale signals to be measured using such instruments?

The keys here are nonlinear response and the stroboscopic technique. An analogous problem was encountered with the development of the very short-pulse lasers we now employ, namely, how can one measure the actual duration of the pulses created? Our usual electronic tools – photodetectors, oscilloscopes – were not (and still are not) nearly fast enough. Fortunately in this case, the problem had already been anticipated, and a solution proposed. The laser pulses can in effect be used as their own yardstick, in the following way. First use a beamsplitter to create two beams from the original beam. Now arrange the beams to cross over in a shallow X, perhaps even focussing each beam to a small diameter at the center of the X. Keep in mind that each beam, although it appears continuous to our eyes, consists of short "bunches" of photons well separated from one another. (From a fairly common and not particularly fast laser, the pulses might be 1 ps in duration recurring at 100 MHz; i.e. the photon bunches span about 300 um from beginning to end, and are separated by 3 m along the beam direction.) Now, if the crossover point is in vacuum, each beam will be unaffected by the presence of the other. Certainly this is the case if the optical path lengths after the beamsplitter differ by more than 300 um. Then the pulses from one beam cross the "empty space" between the pulses in the other. But even if the path lengths are identical, the pulses simply pass through one another, with a linear superposition occurring at the focus.

However, if we place some matter at the focus, the response to the second beam will be influenced by the presence of pulses from the first. Optimizing the choice of nonlinear optical material, we can restrict the response to occur only when there is actual overlap of the pulses in the two beams. A favourite choice is a "second harmonic crystal" in which some light at twice the photon energy of the incident pulses is generated by the nonlinear response. If the response is then measured as a function of the path length difference between the two beams, a profile representative of the actual pulse duration is obtained (the distance axis is converted to time dividing through by the speed of light.) This approach is sometimes called "Armstrong autocorrelation,"[18] and it is the grandfather of all ultrafast optical measurements using two beams known collectively as "pump-probe" techniques. The term "optical boxcar" is also used, after the boxcar signal averager in electronics.

b) Chronology - The first attempt at ultrafast scanning probe microscopy took the nonlinear pump-probe idea and cleverly implemented it with a scanning tunneling microscope. Bob Hamers and Dave Cahill[19] were studying atomic-scale variation of the surface photovoltage of semiconductors. Owing to band bending near the surface, photoexcited electron-hole populations at the surface will experience charge separation, changing the surface potential. The photoresponse is nonlinear (it saturates with increasing illumination), so Hamers and Cahill reasoned the STM could also be used to detect a photovoltage signal in a pump-probe experiment, taking advantage of the nonlinearity of the sample to record the overlap of the probe pulse with response to the pump. Unfortunately the experiment was not successful – one has to worry about thermal expansion of the tip and sample, and other issues affecting the operation of the STM when combined with the laser system. Hamers and Cahill did successfully record time-resolved signals via the tip-sample capacitance when the tip was close to the surface, but not tunneling. The capacitance mode is also useful in scanning probe microscopy, but lacks the fine spatial resolution of STM.

The next implementation of related ideas came with the work of Bloom's group on electrostatic force microscopy (EFM).[20] Here the nonlinear (V^2) force between a conductive force microscope probe and sample is employed to achieve high speed (with pulse biasing) or high frequency measurements of activity in, e.g., integrated circuits.[21] Shortly thereafter the Bridges and Thomson group at Manitoba came out

with their version of this,[22] which they have refined into a quantitative, non-invasive, microscope for mapping electric potentials in high-speed microelectronic circuits. An example of their data is shown in Fig. 5.

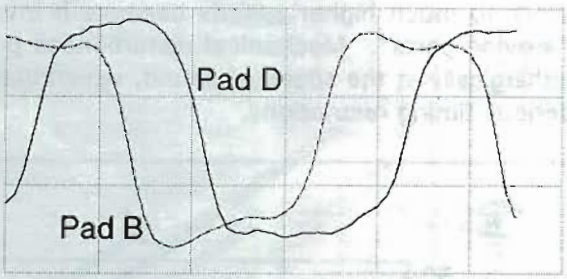

Fig. 5 High-speed electrostatic force microscopy results, courtesy of Profs. G. Bridges and D. Thomson at the University of Manitoba. The data show internal signals in a CMOS circuit operating at 500 Mbit/s (the abscissa is time at 500 ps/div).

Four papers on stroboscopic scanning tunneling microscopy from three different groups appeared within weeks of one another late in 1993.[23-26] Several different approaches had been implemented. In the conceptually simplest method (by the author and G. Nunes, at IBM Yorktown), the distance between the tip and the sample was controlled with a time resolution of tens of nanoseconds.[23] Because of the exponential dependence of the tunneling current on the "width" of the tunnel barrier, the tunneling conductance can be selectively increased for brief intervals by quickly jabbing the tip an angstrom or so closer to the surface. When this is done synchronously with a repetitive process on the sample (e.g. topographic or bias change), a time-resolved measurement with good signal-to-noise can be performed using the STM.

In another method (at Berkeley and Tokyo, called photoconductively gated or PG-STM - refs) the signal from the STM tip was "gated" directly using a fast optoelectronic switch in series with the tip.[24,25] This switch, developed by David Austen (then a Canadian working at Bell Labs) in 1975, is the main key to using the speed of picosecond and femtosecond optical pulses for electronic measurements.[27] Normally open, the switch can be made in such a way that it will close for a duration nearly as short as the optical pulse itself, i.e. only when it is actually illuminated. Such a switch can then be used to generate very fast electrical pulses, or to "sample" fast electrical signals during ultrashort time intervals.

In the PG-STM experiments, an electrical pulse generated by an Austen switch is propagated past an STM tip tunneling into a transmission line. If the other switch in series with the tip is closed at just the right time it should be possible to selectively measure the fast signal, i.e., to time-resolve it (the switch when open is still slightly conductive, but the light can transiently increase the conductivity by many orders of magnitude.) Most of the highest frequency components will be lost if the signal is required to propagate too far up the STM tip, so the "sampling" switch should be as close as possible to the tunnel junction to minimize broadening of the signal before it is gated. An ultrafast tip based on a clever PG-STM tip design of Groenveld is shown in Fig. 6a. The sharp tip is created at the end of a lithographic gold electrode by cleaving the GaAs substrate along perpendicular crystal planes.

Now, a complicating factor in PG-STM (and to a degree in all implementations of ultrafast STM) is the geometric capacitance between the tip and sample. For a typical tip geometry this is in the range of tens of femtofarads. Not a large capacitance, but considering that the signals to be measured may contain frequency components out to hundreds of gigahertz, the peak displacement currents due to the capacitance can be many orders of magnitude larger than the actual peak tunneling current. Furthermore, the "RC" time for discharging this capacitor either through the tunnel junction (typical impedance between 100 MΩ and 1 GΩ) or the open switch should be much too long to measure picosecond pulses. But then, how to explain the very fast signals observed by the PG-STM (as in Fig. 6b)? At first they seem to arise from tunneling because they vanish linearly with the tunneling conductance as the tip is retracted from the surface. It actually took more than a couple of years to resolve this question. Finally Groeneveld showed that, rather than the result of say an anomalously small effective tip-sample capacitance,[28] the very fast signals were in fact capacitive in origin, but manifested in a way that was not apparent until the repetitive nature of the experiment was taken into account. In particular with the gating switch in series with the tip, there is a long enough time constant in the system to carry over some charge from pulse to pulse, and it is not sufficient to calculate the response to a single pump-probe pulse pair. Because PG-STM is a capacitance probe its spatial resolution will not be sufficient for atomic-scale processes, but it is still an interesting technique for high-speed circuit analysis and some

other applications where the "ultimate" resolution is not required.[29] Novel tip designs may yet circumvent this difficulty.[30]

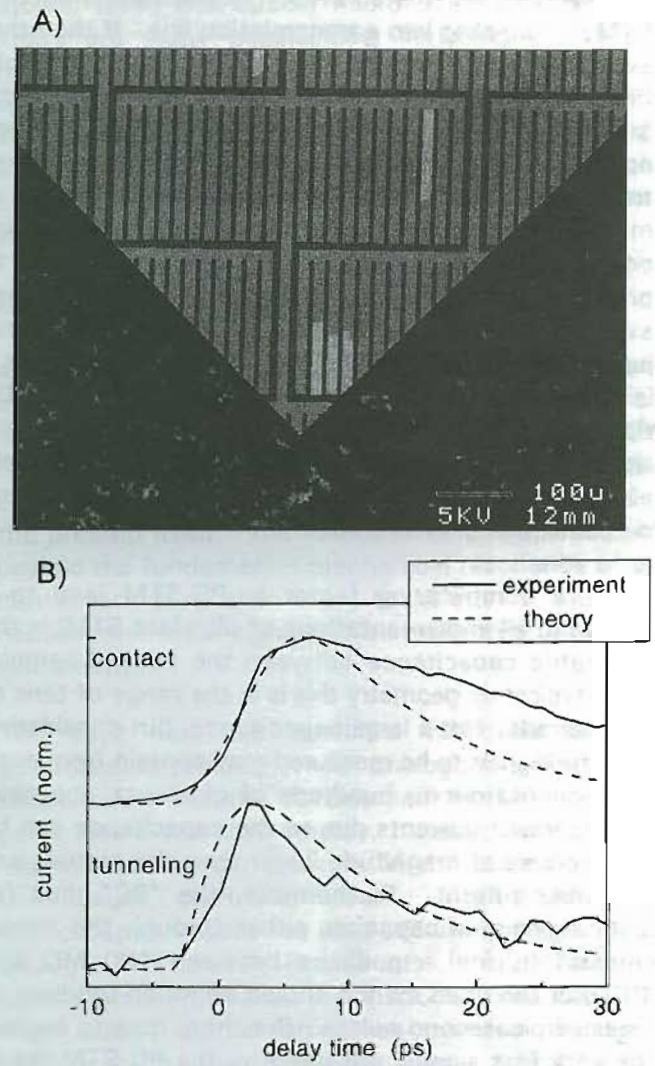

Fig. 6 (a) Electron micrograph of a cleaved STM tip fabricated at the University of Alberta using a modification of the Groeneveld design. (b) University of Nijmegen photoconductively-gated STM data (from Ref. 28) showing subtly different behaviour when the tip is in Ohmic contact with the sample as compared to tunneling. The theory interprets the signals in terms of a capacitance model in this case.

The third approach to ultrafast STM[26] developed simultaneously in 1993 (sometimes called junction-mixed or JM-STM) uses the nonlinearity of the tunnel junction itself to achieve picosecond time resolution, somewhat analogously to the EFM work. Fig. 7 shows the tunneling current as a function of bias voltage (the "current-voltage characteristic") for a PtIr tip tunneling into a gold sample under "ambient" conditions (in air, at room temperature.) Because the slope of this curve increases with increasing bias voltage, we can use transient voltage biasing of the tunnel junction to selectively modulate the effective tunneling conductance (called the differential conductance) for another signal. In a loose analogy, this may be viewed as a variant of the method in which we control the tip-sample separation, but with the advantage that it can work to much higher speeds because it involves no "moving parts". Mechanical disturbances propagate lethargically at the speed of sound, which can impose serious timing restrictions.

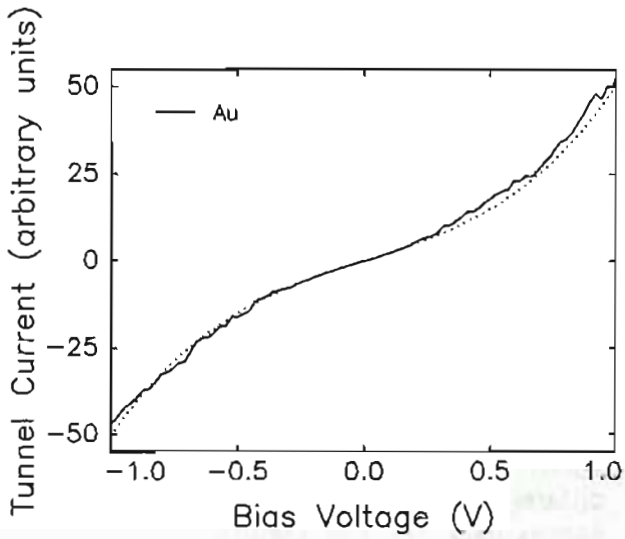

Fig. 7 The current-voltage characteristic for a PtIr tip tunneling into a gold sample under ambient conditions. The solid line is the measurement, the dotted line a fit to determine the (cubic) nonlinear term.

c) JM-STM Details - A diagram of the apparatus used to demonstrate the fastest actual tunnel signal detection to date with an STM is shown in Fig. 8.[31] Two photoconductive switches launch a pair of short electrical pulses onto a transmission line into which the STM is tunneling. The relative timing of the two pulses is again controlled by an optical delay line. One of these pulses is considered as the signal to be measured, while the other acts as the "probe" that, in effect, transiently biases the tunnel junction to a higher differential conductance. To make a "background-free" measurement of the time-resolved signal in this experiment, a "double chopping" technique is used. Both optical beams are chopped (at incommensurate frequencies) and the signal is lock-in

detected at the sum or difference of the chopping frequencies. This way the nonlinear response due to both beams (containing the time-resolved information) is discriminated from the response to each beam separately, which would otherwise appear as a signal independent of the probe delay setting.

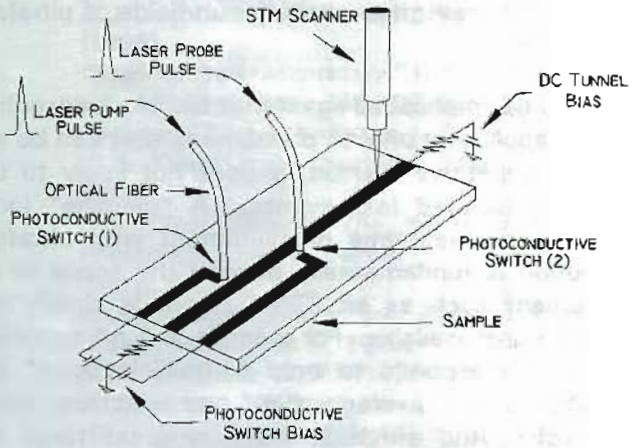

Fig. 8 Schematic of the arrangement used to demonstrate ultrafast scanning tunneling microscopy using the junction mixing technique (from Ref. 31).

A measured ultrafast STM signal is plotted as the solid line in Fig. 9. Ion-implanted GaAs switches were used having a short enough conduction electron and hole lifetime that the electrical pulse widths were essentially limited by the duration of the optical pulses emerging from the optical fibers (about 4 ps). The dashed line is the expected signal as determined by the junction-mixing model, incorporating independent measurements of the I-V characteristic and of the electrical pulse width. The electrical pulse amplitude is the only fitting parameter, and the value of 0.5 V obtained is within the range we expect based on other measurements. Variation of the probe delay at which the signal peak appears as a function of the STM tip position along the transmission line also provides unambiguous evidence that the signal is generated at the tip. There is no observable contribution from other nonlinearities, such as an interaction between the two photoconductive switches, which conceivably could influence the signal.

And what is the role of the geometric capacitance in the JM-STM experiments? As the signal is generated entirely by a tunneling current, the tip-sample capacitance should not degrade the spatial resolution in JM-STM measurements. It will place a cap on the ultimate temporal resolution, however. As the electrical pulse widths become shorter (and the characteristic frequencies they contain become proportionally higher), the electromagnetic coupling between the tip and the sample becomes stronger (governed by the geometric capacitance in a lumped circuit element model.) Eventually with increasing speed, the transient potential difference induced between tip and sample will be substantially reduced, and the probe pulse will no longer be effective.

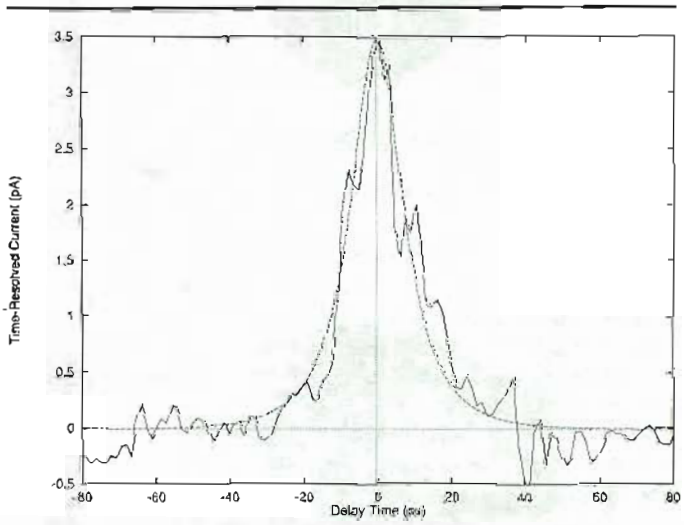

Fig. 9 A short electrical pulse measured using the JM-STM (dashed line). The solid line is from a calculation of the expected signal.

In order to perform a preliminary demonstration of spatial resolution in junction-mixing STM, we have artificially patterned a surface using two different metals to produce a spatial variation of the tunneling nonlinearity. A time-resolved signal measured as a function of position then reflects this spatial dependence. Fig. 10 shows data from a structure consisting of 30 nm-thick titanium squares patterned on a gold surface. A spatial resolution of better than 20 nm is achieved in the stroboscopic signal, probably limited by the lithographic resolution of the squares themselves. More refined measurements are in progress, and we believe that atomic resolution will be possible using the technique.

DISCUSSION

The capabilities for ultrafast scanned probe microscopy in general will continue to expand for the forseeable future. This is all but guaranteed by progress in the component systems such as lasers and micromachined probes, but will also increasingly be fueled by new and developing applications across the physical and life sciences and engineering. Electronic and magnetic devices in the sub-micrometer range are quickly

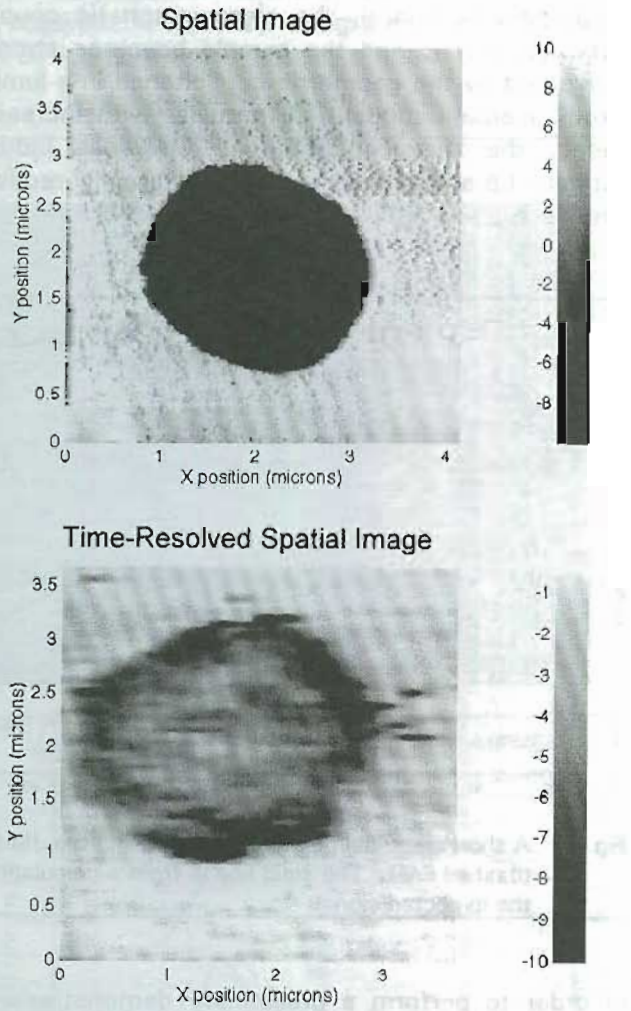

Fig. 10 Images from an experiment designed to begin to characterize the spatial resolution achieved in practice using the JM-STM technique for picosecond measurements. (a) Conventional STM image of a thin film titanium dot patterned on a gold surface. (b) A time-resolved image from such a structure. In short scans across an edge the spatial resolution is found to be clearly less than 20 nm (which in this case is approximately the thickness of the dot.)

gaining in importance, and the dynamics of such small systems are invariably fast. The range of biological processes to be explored is enormous, although here the challenge is greater because the samples are less robust. Every significant improvement in combined spatial and temporal resolution opens new opportunities.

As for the ultimate goal of combined "atomic" spatial and temporal resolution, a great deal of exciting and challenging development remains to be done. The present implementation of JM-STM using photoconductively switched electrical probe pulses will run out of steam somewhere between 100 fs and 1 ps. Two difficulties will be encountered in this range, an inability to increase the speed of the Austen switches any further, and the effect of the tip-sample capacitance. Further progress may come through the use of electromagnetic pulses propagated through free space then coupled to the tip of the STM. Methods for generating and delivering such pulses, with a wide range of carrier frequencies from terahertz to optical, already underlay other exciting sub-fields of ultrafast studies.[32]

It should be emphasized again that the work described here is applicable only to phenomena that can be set up in a repetitive fashion; it does not apply to the study of isolated fast events. A "genuine" (non-stroboscopic) real-time measurement with ultrafast resolution is fundamentally beyond the scope of an instrument such as an STM: there is simply not enough signal available. For example, a 1 nA tunneling current corresponds to only 6 "millielectrons" per picosecond on average (i.e. one electron every 160 ps). And although there is great room for development of ultrafast "instantaneous" snapshots using otherwise conventional optical imaging methods, such approaches are fundamentally incompatible with the raster-scanned image acquisition mode of a high resolution SPM.

Many exciting studies await all of these developments. The near- and intermediate-term scientific interests motivating our current work include nonequilibrium dynamics in micrometer- to nanometer-scale superconductors and magnets. Direct, time-resolved scanning probe microscopy seems an essential tool in the elucidation of these systems where very fine-scale features and nonlocal interactions influence both the static structure and the dynamics. Interest is heightened by the opportunity to study long-standing scientific problems such as magnetization reversal, by the potential for these systems to serve as laboratory models for other fundamental physical processes, and by the connection to practical applications.[33] Ultimately the work shares much in common with the animal locomotion studies of Muybridge in the 19th century. We've simply moved on from "too fast for the unaided eye to distinguish" to "*way* too fast *and* too small."

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    Ultrafast illumination is still expensive, but getting cheaper all the time. Amortizing the cost of a typical, popular Ti:sapphire system, photons in femtosecond pulses can cost about $1000/Einstein, where an Einstein is a mole of photons. Hopefully within ten years the femtosecond equivalent of continuous-wave laser pointers will make it possible to show real ultrafast phenomena as lecture demos!
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