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# Ultrafast Scanning Microscopy

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## Abstract
We propose to combine ultrafast optical techniques with scanning probe microscopies to obtain, simultaneously, unprecedented temporal and spatial-resolution.

## Introduction
Very high spatial resolution is required for investigating the phenomena that govern the physics of mesoscopic systems and for characterizing the operation of submicron optoelectronics devices. For example, processes such as: i) carrier transport in mesoscopic semiconductor structures; ii) electric field and voltage wave front propagation at metal-semiconductor interfaces; iii) light emission in quantum confined structures; all experience significant variations over length scales much smaller than 1μm. Because of the large velocities of excitations in the materials (electronic velocity in semiconductors and metals is on the order of ≈ 1 to 10Å/fs; voltage wave fronts propagate on high speed transmission lines at velocities on the order of ≈ 1000Å/fs), the time scale of their dynamics is well in the sub-picosecond domain. Therefore, in order to understand better the mechanisms governing mesoscopic systems and optimize the devices that use them, it is necessary to develop new experimental techniques with **simultaneous** subpicosecond-time resolution and submicron-space resolutions.

In the last decade, powerful scanning microscopy techniques which can achieve atomic resolution have been developed. The invention of the Scanning Tunneling Microscope (STM) [2], the Scanning Force Microscope (SFM) [3] and the Near-field Scanning Optical Microscope (NSOM) [4] have revolutionized the field of surface science. In the scanning microscopies an atomically sharp tip (or a very small aperture for the NSOM) is scanned very close to the surface of a sample. Due to the close proximity of the tip to the sample, different interactions or coupling mechanisms can occur (tunneling current, atomic forces, evanescent wave coupling). The strength of these interactions is a very nonlinear function of the tip height. This nonlinearity provides the means for atomic-scale control of the distance to the sample and allows high lateral resolution limited by the sharpness of the tip. In STM, the image, obtained by collecting the tunneling current while scanning the tip, reflects the density of states of the electrons at the Fermi level close to the surface. In SFM, the image is obtained by measuring the minute deflection of the cantilever holding the tip, caused by the force exerted on the latter by the sample (electrostatic and magnetostatic forces, inter-atomic forces, van der Waals forces etc.). The NSOM image is obtained by evanescent field coupling to a sub-wavelength aperture. Presently, however, the time-resolution of the scanning microscopies is limited by the scanning rate and the data acquisition electronics.

## Basic Concept
We propose to exploit the nonlinear nature of the different tip-sample interactions to obtain ultrafast time-resolution in scanning microscopy. This nonlinearity can be intrinsic to the nature of tip-sample coupling or can be artificially produced by **nano-fabrication of costume designed tips.** The time

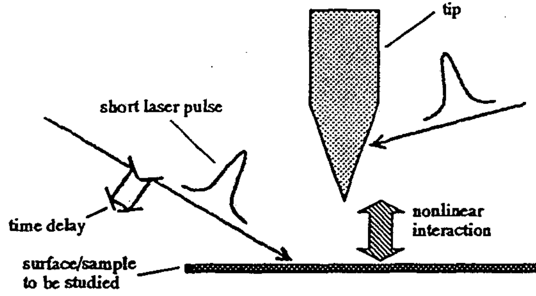


**Fig. 1:** Correlation by nonlinear interaction. The two laser pulses modulate the sample and the tip responses respectively.

resolution is achieved by modulating both the sample and the tip responses with two short optical pulses, as shown in Fig. 1. Because of the nonlinearities, multiplicative ("mixing") terms are generated, which include both tip and sample responses. By scanning the time delay between the two pulses and integrating the signal, a cross-correlation of the tip-sample responses is obtained. After characterizing the response of the tip alone, the dynamics of the sample can be obtained by deconvolution, with the simultaneous spatial and temporal resolutions which are well below 1μm and on the order of 1ps respectively.

This basic principle is very general, and can be implemented in wide variety of applications. In the following we describe and discuss two examples.

## Ultrafast STM

An example of artificially induced nonlinearity is shown in Fig. 2, where an atomically-sharp tip is integrated with an ultrafast photoconductive-switch [5]. In this experiment, we plan to study electromagnetic wave propagation and distribution of the field lines on a transmission line with sub-micron and sub-picosecond resolutions. This experiment will

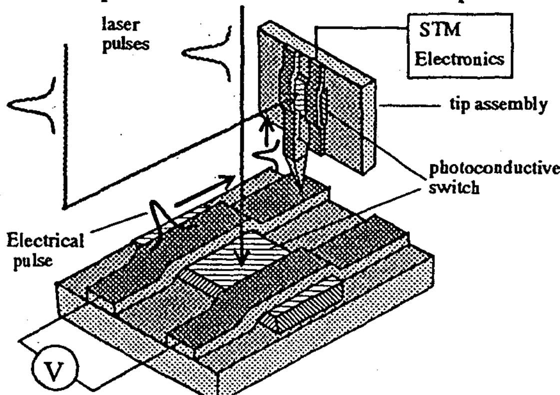


**Fig. 2:** Ultrafast STM. One laser pulse excites a voltage pulse on a transmission line. The second pulse photoconductively samples the tunneling current on the tip assembly.

also determine the δ-function response (to a voltage pulse) of the system. In the figure, one pulse excites the line or a device integrated in the line, while the second pulse samples the tunneling current on the tip assembly.

In order to estimate the signal contrast in such an experiment, we use the equivalent electrical circuit

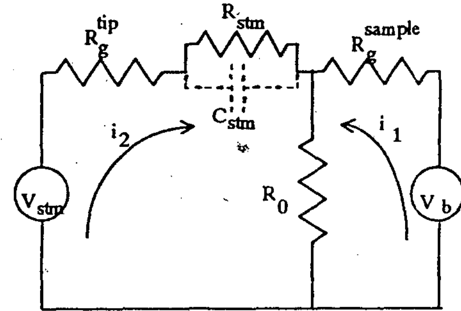


**Fig. 3:** Equivalent circuit for Fig. 2. The STM gap is modeled as a resistor in parallel with a capacitor.

which is shown in Fig. 3, and calculate onlt the DC component of the signal. The transmission lines are modeled by a resistor $R_0 = 100\Omega$, the photo-conducting gaps as resistors with variable resistance (light on: $R_g^{tip} = R_g^{sample} = 1M\Omega$ , light off: $R_g^{tip} = R_g^{sample} = 10M\Omega$), the tunneling gap has a resistance of $R_{stm} = 10M\Omega$ and a capacitance $C_{stm}$ (which we neglect for the DC analysis). We assume $V_{stm} = 10mv$ and $V_b = 10v$. These numbers roughly correspond to the parameters of LT-GaAs (i.e. GaAs epitaxially grown at low temperature). The DC component of the tunneling current is given by:

$$i_2 \cong \frac{V_{stm} - V_b \dfrac{R_0}{R_g^{sample}}}{R_g^{tip} + R_{stm}} \qquad (1)$$

With a duty cycle of $10^{-4}$, which corresponds to a 100 MHz repetition rate of the laser source and 1ps time resolution, we get $i_2^{light\,on} / i_2^{light\,off} \cong 2$ with 0.5 nA nominal tunneling current (assuming lock-in detection). This simple calculation takes into account only the DC coupling of the terahertz pulse to the tip. We suspect that AC coupling will play an important role. The AC coupling, though is very difficult to model for the proposed geometry, and further studies will be needed to clarify its contribution to the signal.

We have performed a simulation experiment, in a planar geometry, using silicon on sapphire (SOS) as the photoconductive material. It turns out that the resistance of the silicon gap (≈10 to 20 μm) is

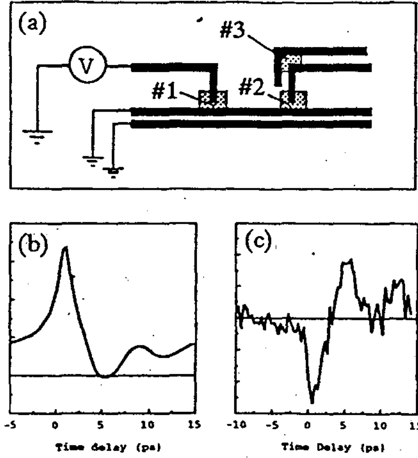


**Fig. 4:** Simulation experiment on SOS. (a) mask layout and biasing geometry (b) conventional photoconductive sampling (points #1 and #2) (c) photoconductive sampling between points #1 and #3.

insulator/vacuum - metal). Fig. 4a shows the mask layout and the biasing. One laser beam switched the bias at point #1. The second beam switched either point #2 (conventional geometry for photoconductive sampling) or point #3. The later geometry simulates the STM experiment. The resulted signals are shown in Fig. 4b and 4c respectively. The amplitude of the signal in Fig. 4c is multiplied by a factor of $10^3$. The corresponding sampled current is $\approx$10pA. As can be seen, the signal in Fig. 4c has a strong AC feature, which indicates capacitive coupling.

## Ultrafast SFM

Another utilization of the basic concept is the use of the SFM as the nonlinear cross-correlator. With two

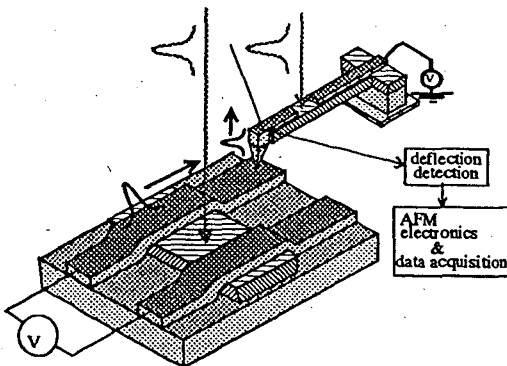


**Fig. 5:** Ultrafast SFM. One laser pulse excites a pulse on a transmission line. The second pulse modulates the voltage of the SFM tip.

short optical pulses, one can modulate the force acting on the SFM tip. In the attractive mode, the SFM can measure the electrostatic force (Coulomb interaction) exerted on the tip by the electrical charge/voltage deposited on the sample. Fig. 5 shows a tip mounted on a cantilever which is micro-machined from a photoconducting substrate. The tip voltage is modulated by a short optical pulse. Its deflection is measured by optical techniques. The second short laser pulse excites the device. The cantilever acts as a slow detector which cross-correlates the dynamic responses of the sample and the tip. Its deflection (as function of time delay between the pulses) provides information on the dynamics of the device.

The electrostatic force acting on the tip can be modeled as the force acting on a plane-parallel capacitor. The equation of motion for the tip-lever is given by:

$$m\ddot{x} + \gamma\dot{x} + k\,x = \frac{\varepsilon_0\,A}{U_0{}^2}V(t)^2\left(1 - \frac{2x}{U_0} + \frac{3x^2}{U_0^2}\cdots\cdots\right) \quad (2)$$

where x is the deviation of the tip from its equilibrium position, m is the mass of the lever, $\gamma$ is its dissipation, k is its spring constant, $\varepsilon_0$ is the vacuum permittivity, A is the area of the tip, V is the tip-sample voltage and $U_0$ is the equilibrium position. The differential force, with respect to the tip height, is given by:

$$\frac{\partial F}{\partial z} = \varepsilon_0\,\frac{A}{z^3}\,V^2. \quad (3)$$

With an average change in voltage of 1mV, nominal tip height of 100 Å, and a tip radius of 1 µm (which, of course, affects the resolution), a gradient force of $10^{-5}$ N/m can be obtained. Force gradients of that order of magnitude have been measured successfully with a SFM.

To demonstrate how the SFM acts as a correlator, we solve (2) with the following excitation (train of δ-function voltage pulses):

$$V_t(t) = \sum_{n=0}^{N}\delta(t - nT)\ ,$$

$$V_s(t-\tau) = \sum_{n=0}^{N}\delta(t-\tau - nT) \quad (4)$$

T is the separation between adjacent pulses and N·T is the time of integration on the detector. The tip-sample bias is: $V(t) = V_t(t)+V_S(t-\tau)$. The 0-order solution (we take only the first term in the expansion in (2)) is given by:

$$x(t) = \sum_{n=0}^{N} x_\delta (t-nT) + \sum_{n=0}^{N} x_\delta (t-\tau-nT) + \begin{cases} 2\sum_{n=0}^{N} x_\delta (t-nT) & \tau = 0 \\ 0 & \tau \neq 0 \end{cases} \tag{5}$$

with:

$$x_\delta(t) = \left( C_1 - \frac{1}{\sqrt{\gamma^2 - 4km}} \Theta(t) \right) e^{-\left(\frac{\gamma + \sqrt{\gamma^2-4km}}{2m}\right)t} + \left( C_2 + \frac{1}{\sqrt{\gamma^2 - 4km}} \Theta(t) \right) e^{-\left(\frac{\gamma - \sqrt{\gamma^2-4km}}{2m}\right)t} . \tag{6}$$

As can be seen from (5) and (6), the tip deviation at $\tau=0$ is twice as big as the deviation at $\tau\neq0$. This calculation is ofcourse oversimplified, but it has all the basic ingredients to demonstrate the basic concept brought-up in this proposal.

## Conclusions

We propose to combine ultrafast techniques with scanning microscopy techniques. The development of such instruments will open the way for a new kind of spectroscopy with the **simultaneous** subpicosecond-time and submicron-space resolutions.

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