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Progress in Ultrafast Scanning Probe Microscopy

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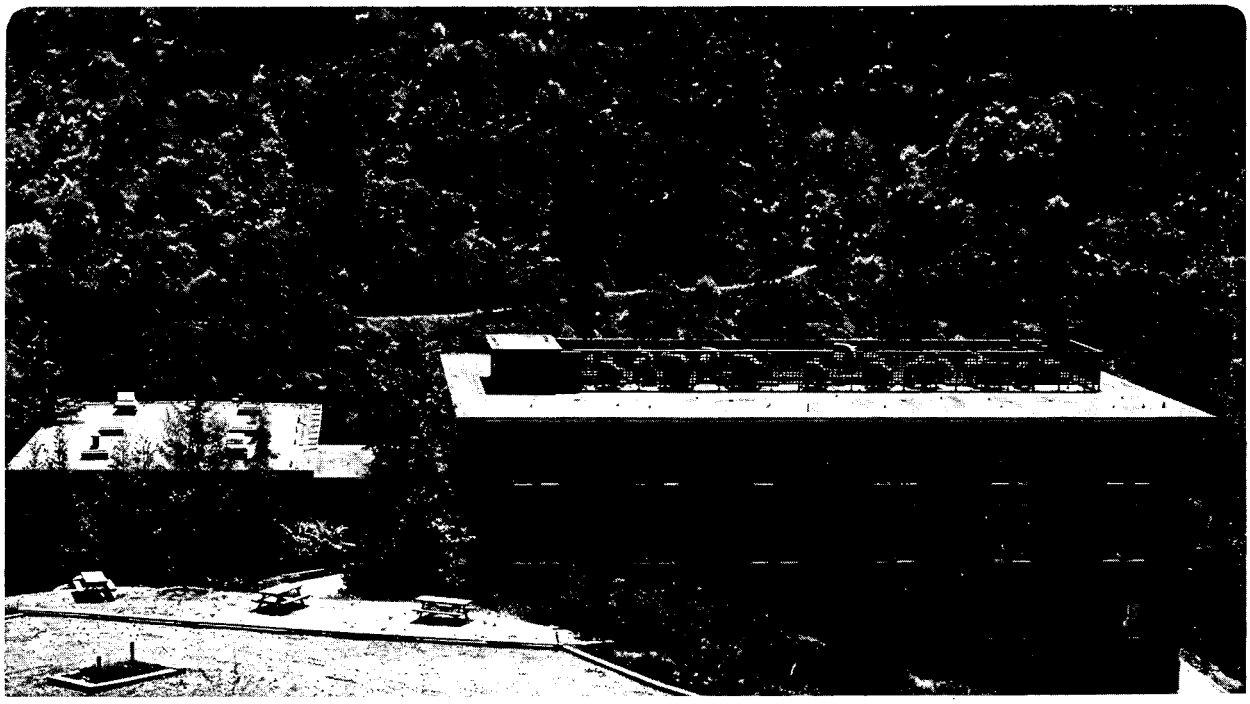

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PROGRESS IN ULTRAFAST SCANNING PROBE MICROSCOPY

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Abstract

We demonstrate simultaneous picosecond time and atomic space resolution measurements. The room temperature quantum mechanical capacitance of the STM point contact is observed for the first time. The potential for ultrafast movies of surface dynamics on atomic scale is discussed.

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Recently, we proposed a general technique to wed ultrafast laser spectroscopy and Scanning Probe Microscopies (SPM) to obtain simultaneous picosecond time and atomic space resolution[1]. Demonstration of this concept immediately followed.[2] In the experiment, a short (100 fs) laser pulse activated a photoconductive switch on a SOS transmission line, in order to generate short (650 fs) voltage pulses. While the tip was in tunneling range from one of the conductors of the transmission line, a second laser pulse gated the Scanning Tunnelng Microscope (STM) tip assembly. Thus, we could excite the tunneling gap of the STM with a short voltage pulse, and measure its response. We also used this instrument to image a surface with 2 ps time resolution and better than 50 Å spatial resolution. The details of the experimental arrangement are described eleswhere.[2]

A typical signal measured by the STM consists of a small AC component which varies with time delay riding on a large DC background. The background is simply the nominal tuneling current, given by (bias voltage)/(gap resistance). Fig 1a shows a 4.5 ps wide cross-correlation pulse detected by the STM while tunneling at 5 nA with a +80 mV bias on the strip. The 10% to 90% rise time is 2 ps. The average tunnel current is increased by ~0.25 nA when the transmission line pulse passes beneath the tip at zero time delay. This increase corresponds to a signal/background ratio, DI/I, of 5%. Fig. 1b shows a cross-correlation recorded while the tip was in ohmic contact with the sample. The bipolar correlation peak in Fig. 1a is significantly different from what we would expect for a purely ohmic response. It bears some resemblance to Fig. 1c, which is the numerical derivative of the trace in Fig. 1b. Apparently, a large portion of the tunneling signal comes from a capacitive-like response. However, the tunneling gap height dependence of this capacitive-like response is very different from the one expected for the geometrical capacitance of the junction. In fact; the geometrical capacitance of the tip assembly contributes negligibly to the signal.[2]

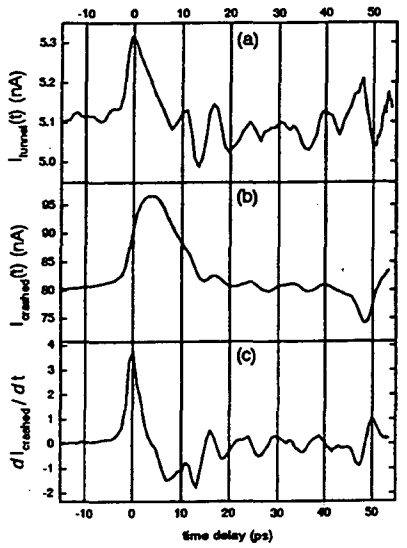

Figure 1. Time resolved current cross-correlation detected on the tip: (a) in tunneling range (5nA and 80mV settings), and (b) when the tip is crashed into the sample. (c) is the time derivative of (b).

To better understand these measurements, we model the experiment by an equivalent circuit[3] in which the tunnel junction impedance is described by a resistor R in parallel to a capacitor C. We find that the time resolved tunnel current I_t can be determined, to within a single fitting parameter, by the ohmic contact measurment I_{cr}:

$$I_t = a \cdot (I_{cr} + \tau \cdot \frac{dI_{cr}}{dt}) \qquad (1)$$

a is a normalization constant, and $\tau = RC$ is the fitting parameter (in units of picosecond). Fig. 2 shows such a fit, obtained for the trace shown in Fig. 1a with the trace of Fig. 1b. Our model does not account for the high frequency, oscillatory part of the signal. As can be seen from the figure, the envelope of the tunneling data is fitted very well. In particular, the rise and fall times of the signal at time delay 0 are nicely matched. Since R is

measured by the STM electronics, we can extract the capacitance of the junction: $C = \tau / R$. From such fits, done on various data sets, we find that the capacitance of the junction is on the order of $10^{-20\pm1}$ F. This capacitance is much smaller than the claculated geometrical capacitance (on the order of fF). Using the same procedure, we fitted a series of data of varying tip height. The results of these fits are shown in Fig. 3, where we plot C as function of $G = 1/R$, the tunneling gap conductance. We find a linear relation between the two. Since the gap conductance is exponential with tip height, we conclude that the capacitance has the same height dependence. Both the small magnitude and the height dependence suggests that the origin of the observed capacitance is quantum mechanical.

Quantum mechanical susceptance, and in particular, quantum capacitance, associated with the tunneling process itself, has been discussed before for the Josephson effect. Both the Cooper pair tunneling and the quasiparticle current tunneling display quantum capacitance at high frequencies. The susceptance associated with the Cooper pair tunneling originates from the phase difference between wave functions on both sides of the gap. For the quasiparticle current case, "quantum sloshing" is responsible for the suceptance[4]. Both phenomena require sharp density of states (the superconducting gap) and very low temperature. In our experiment, we observe quantum mechnical capacitance between two normal metals, at room temperature. We are currntly contrasting our results with the photon assisted tunneling theory[4] to check if it can explain our experimental observations.

One of the most exciting prospects for USPM is the potential to create movies of surface dynamics. We believe that by improving the sensitivity, dynamic range, and time resolution of this technique, we will be able to animate surface dynamics by collecting a series of STM images for increasing values of time delay. This will allow us to study dynamical phenomena in real space with atomic resolution. Such a tool will open a window for the observation of processes and excitations which propagate at velocities of a few Å per fs (or less). It should be possible to spatially resolve in real time phenomena such as vibronic motion on the atomic scale, carrier transport in semiconductor structures, electric field and voltage wavefront propagation at metal semiconductor interfaces. The operation of sub-micron electronic/optoelectronic devices can be directly characterized with such a technique.

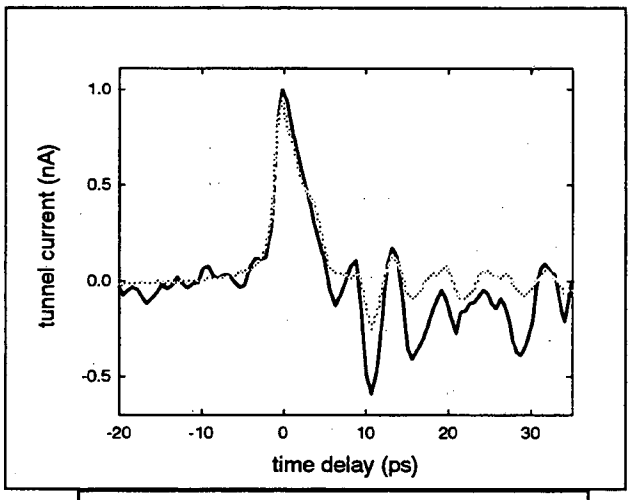

Fig. 2: Fit (solid gray line) to the time resolved data (solid black line) with the ohmic contact signal

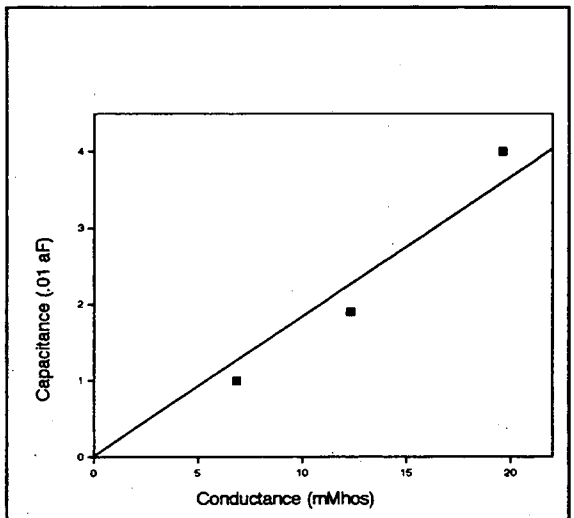

Fig. 3: Tunnel gap capacitance vs. conductance (the line is a guide to the eye)

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References

[1]S. Weiss, D. Botkin and D.S. Chemla, OSA proceedings on *Ultrafast Electronics & Optoelectronics*, vol.14, San Francisco, January 25 1993, edited by J. Shah and U. Mishra, (OSA, Washington D.C. 1993), pp.162-165.

[2]S. Weiss, D.F. Ogletree, D. Botkin M. Salmeron and D.S. Chemla, Appl. Phys. Lett., **63**, 2567 (1993).

[3]S. Weiss, D. Botkin, D.F. Ogletree, M. Salmeron and D.S. Chemla, to be published.

[4]J.R. Tucker and M.J. Feldman, Rev. Mod. Phys., **57**, 1055 (1985) and references cited therin.

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