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ADVANCES IN ULTRAFAST SCANNING PROBE MICROSCOPY

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Ultrafast 94

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Abstract

We review the development of the ultrafast scanning tunneling microscope. Experimental results on the tunneling gap response to a short voltage pulse excitation are presented.

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Recently, we have 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: we have implemented this general technique in a specific device, the Ultrafast Scanning Tunnelng Microscope (USTM).[2] Ultrafast time response occurs when the SPM probe has a nonlinear response to a short laser pulse. This nonlinearity can be essential to the nature of the probe-sample interaction or can be artificially introduced to the probe. For instance, the intrinsic nonlinearity of the STM I-V characteristic can be used to obtain ultrafast time resolution.[1,3] The tip nonlinearity mixes the high frequency (short time) responses to produce a DC (correlation) signal. Temporal resolution is defined by the temporal width of the nonlinear tip response and the width of the laser pulses. Spatial resolution should be comparable to the resolution of the normal SPM. In the first demonstration, the nonlinear element was a fast photoconductive switch integrated with the STM tip assembly. Conceptually, the switch acts as a several picoseconds long gate.

In the experiment, the sample was an Au transmission line. A short (100 fs) laser pulse switched a photoconductive switch on the sample, to generate short (650 fs) voltage pulses on the line. A second laser pulse gated the Scanning Tunnelng Microscope (STM) tip. Thus, we could excite the tunneling gap of the STM with a short voltage pulse, and measure its response. We used this instrument to image surfaces with 2 ps time resolution and better than 50 Å spatial resolution. The details of the experimental set-up are described eleswhere.[2]

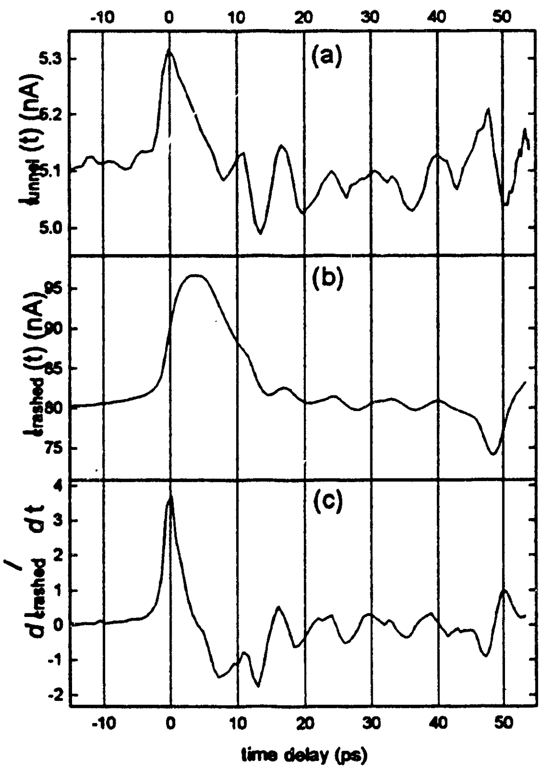

Figure 1. Time resolved current cross-correlation detected on the tip: (a) in tunneling range (5nA and 80mV settings), and (b) when the tip is crashed into the sample. (c) is the time derivative of (b).

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.

There are several features in this data. The large signal at a time delay of 47 ps is a reflection from one end of the sample transmission line. This reflection also appeared in our standard sampling measurements. The structure following the main correlation peak is due to ringing in the tip structure, as we will show below. The FWHM of the correlation peak in Fig. 1b is much broader (10 ps) than the pulse on the transmission line. The broadening is, most likely, a result of dispersion of the electrical pulse on the tip. The bipolar correlation peak in Fig. 1a is significantly different from what we would expect for a purely ohmic response; in fact 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; as we will show below, the geometrical capacitance of the tip assembly contributes negligibly to the signal. The existance of an intrinsic capacitance associated with the tunneling process has been discussed in the context of Coulomb Blockade (CB) and Single Electron Tunneling (SET).[4] This capacitance is beleived to be very small (on the order of 10^{-18} F). The RC discharge time for such a capacitance at reasonable tunneling gap resistances is on the order of a few ps and can be directly investigated with our USTM.

Fig. 2a displays a series of cross-correlation pulses recorded by the STM while tunneling with a +80 mV bias on the strip for gap resistances from 16 MW to 512 MW. For clarity, the DC background in each curve has been removed. In Fig. 2b, we normalize the peak height of the traces in Fig. 2a to unity in order to show changes in the lineshape of the signal. The figure shows that the fall time of the correlations increases with increasing gap resistance. This increase may be associated with the RC discharge time discussed above.

The spectra of the traces in Fig. 2a are shown in Fig. 2c. These spectra were calculated for data in the range -10 ps to 37 ps. Additionally, the heights of the low frequency peaks have been normalized to 1. The low frequency peak describes the main bipolar correlation peak. The smaller high frequency peaks represent the ringing effect in the correlation traces. This ringing is probably due to an impedance mismatch between the tip and the sample and between the tip and the sampling switch. As the tunneling resistance is reduced, the mismatch is reduced, and the amplitude of the ringing peak is reduced. The fine structure in the ringing has not been identified.

The height of the correlation peak in each trace, as function of the corresponding DC current is shown in Fig. 3. The filled squares represent measurements at 16, 32, 64, 128, 256 and 512 MW

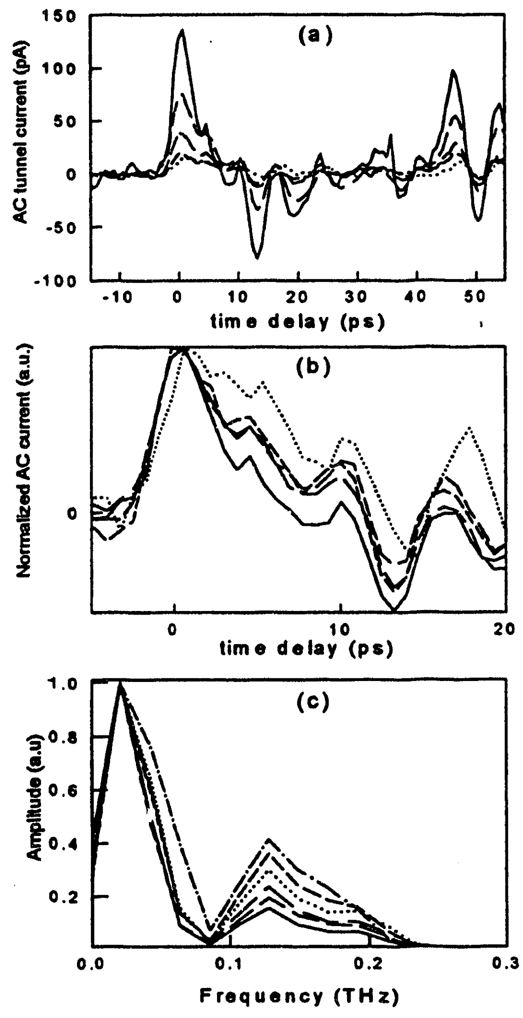

Figure 2. a) The tunnel-current DI(Dt) for different gap resistances. (solid line is 16 MW, long dash - 32 MW, medium dash - 64 MW, short dash - 128 MW, dotted- 256 MW, dash-dot - 512 MW). b) Traces in a) with peak heights normalized to unity. c) Spectra of the curves in a).

gap resistances (the point at the top right of the figure corresponds to 16MW). The dashed line is a linear regression to the data. It is clear from the figure that the AC part of the tunneling current has the same gap impedance, and hence distance, dependence as that of the DC current. Moreover, when the tip is withdrawn from the surface by 50 Å, not only does the DC tunnel current vanish (as expected), but the AC part nearly vanishes as well. This means that the observed cross-correlation signal has little or no contribution from stray capacitance in the leads or from radiative coupling.

A detailed study of the nature and the origin of the tunnel-gap capacitance is currently under way. By varying the tip length, the gap conditions (by controlling the oxide layer) and the bias voltage on the transmission line, we will separate the contribution of the stray capacitance of the leads and reflections along the tip from the intrinsic response of the tunneling junction itself. We plan to improve the time resolution of our USTM by microfabricating the entire tip-switch assembly.

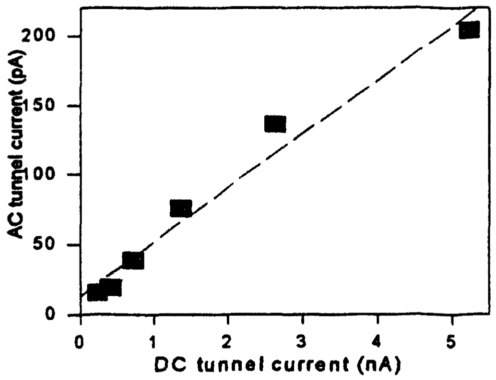

Figure 3. Amplitude of the time resolved signal versus DC background. The dotted line is a linear regression to the data.

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.

USPM is not limited to the STM. As we discussed before,[1] the non-linear nature of the probe-sample interactions in STM, AFM and near field optical microscopy (NSOM) offer many opportunities for resolving ultrafast phenomena on a nanometer scale.

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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]G. Nunes and M.R. Freeman, Science, 262, 1029 (1993).

[4]For example, K.K Likharev, IBM J. Res. Develop., 32, 144 (1988).

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