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**ULTRAFAST SCANNING PROBE MICROSCOPY:
TOWARDS ULTRAFAST MOVIES OF MOVING ATOMS**

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***Ultrafast Scanning Probe Microscopy:
Towards Ultrafast Movies of Moving Atoms***

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Abstract

We have merged the techniques of ultrafast laser spectroscopy with those of scanning probe microscopy to develop an ultrafast scanning tunneling microscope. The response of a tunneling gap to a 650 fs voltage pulse was measured.

Ultrafast Scanning Probe Microscopy: Towards Ultrafast Movies of Moving Atoms

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By combining ultrashort laser pulse techniques with scanning tunneling microscopy (STM), we have developed an instrument which obtains simultaneous 2 ps time resolution and 50 Å spatial resolution. This is a nine orders of magnitude improvement over the time resolution previously attainable with STM. We have used this instrument to measure the response of the tunneling gap to excitation by a sub picosecond electrical pulse. Our technique is not limited to STM; it can be implemented in a variety of scanning probe microscopies, allowing the observation of ultrafast dynamics on the atomic scale.

The nonlinear tip-sample interaction in scanning probe microscopy (SPM) enables one to measure laser induced dynamical events on surfaces. This is done by adapting the standard pump-probe technique to SPM.¹ The first demonstration of this concept utilized a photoconductive gate on the STM tip and a photoconductively generated voltage pulse on the sample.^{2,3}

In the experiment (Fig. 1), 100 fs pulses from a mode locked Ti:sapphire laser photoconductively excited ~650 fs wide voltage pulses on the sample transmission line. As those voltage pulses passed under the STM tip, a second laser beam gated the switch on the tip assembly. The tunnel current was recorded as a function of time delay between the two laser pulses. In Fig. 2 we show a series of time-resolved tunnel current correlations, taken at different tip-sample separations. For clarity, only the changes in the tunnel current (the AC response) are shown. 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 MΩ gap resistances (the point at the top

left of the figure corresponds to $16\text{M}\Omega$). The dashed line is a linear regression to the data. It is clear from the Fig. that the AC part of the tunneling current has the same gap impedance (and distance) dependence as that of the DC current. Moreover, when the tip is withdrawn from the surface by 50 \AA , 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.

We also took STM images of the transmission line for different time delays. By collecting a series of such STM images for increasing values of time delays, we expect to be able to "produce" ultrafast movies on the atomic scale. The technique will be a powerful new tool for the observation of processes and excitations which propagate at velocities of a few \AA per fs. We believe that it will be possible to spatially and temporally resolve many dynamic phenomena on an atomic scale. Future investigations will focus on vibronic motion of atoms on surfaces; carrier transport in semiconductors, molecules and semiconductor devices; and hot carrier effects.

Acknowledgments

This work was supported by the Laboratory Directed Research and Development Program of Lawrence Berkeley Laboratory under the U.S. Department of Energy, Contract No. DE-AC03-76SF00098 and by ONR/ARPA under Contract No. N000-14.93.105.36.

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Figure Caption:

Fig. 1: experimental set-up: One laser pulse excites a voltage pulse on a transmission line.

The second pulse photoconductively samples the tunneling current on the tip assembly.

Fig. 2: Time resolved tunnel-current for different gap resistances (16, 32, 64, 128 and 256M Ω from top to bottom)

Fig. 3: The AC tunnel current plotted vs. the DC tunnel current for different gap resistances (16, 32, 64, 128, 256 and 512 M Ω).

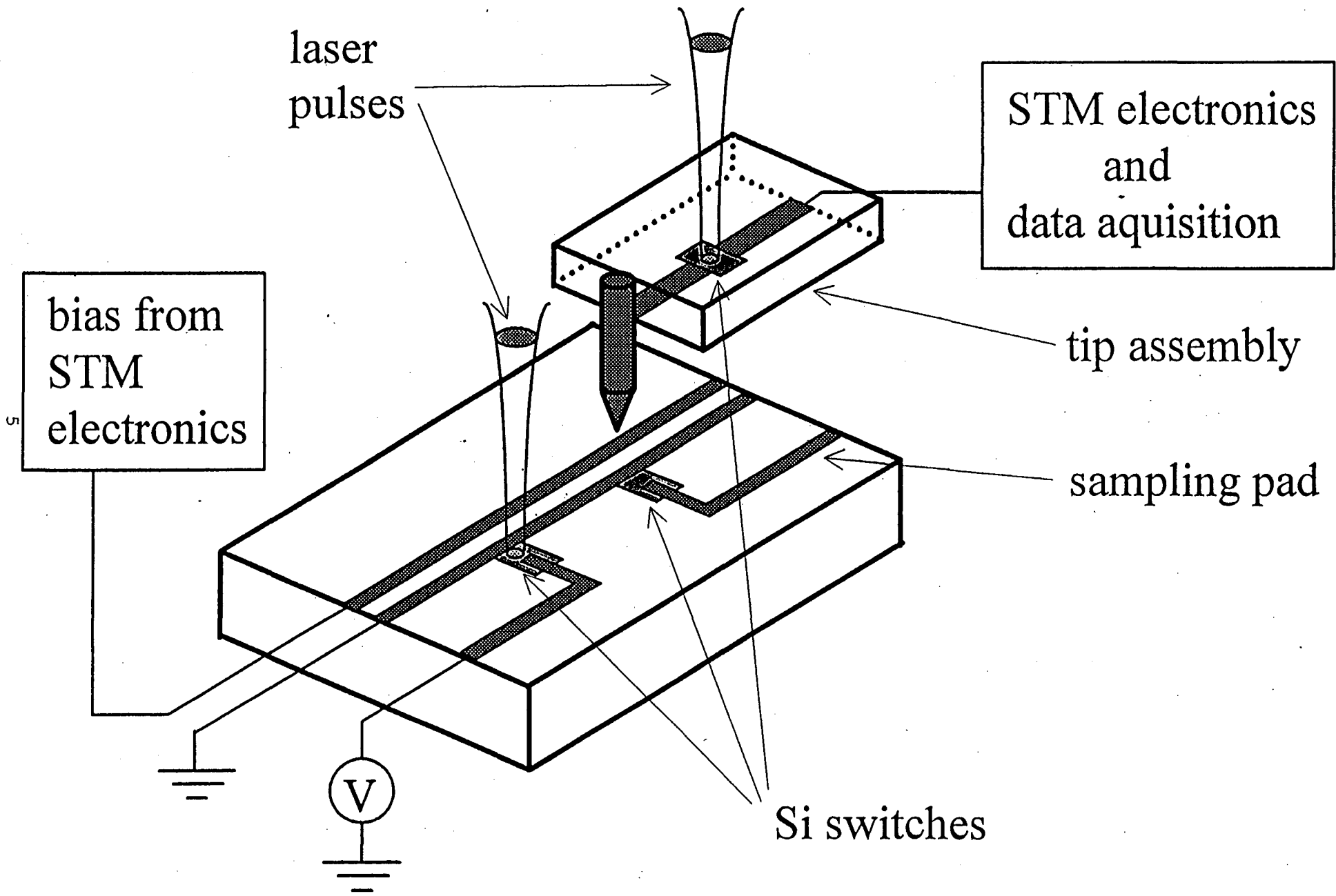


Fig 1
 Schematic diagram of the STM system

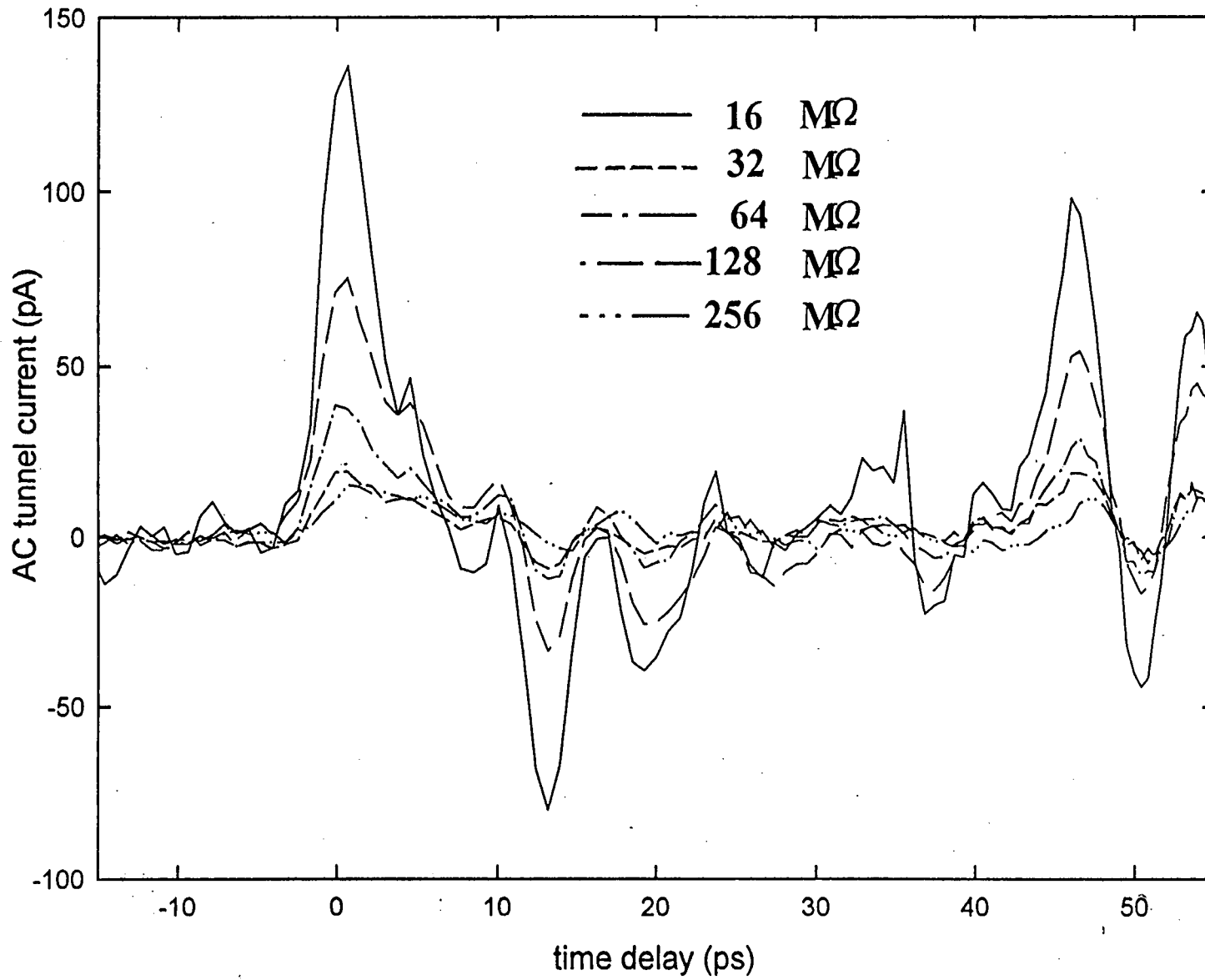


Fig. 2

Weiss et. al.: "ultrafast scanning..."

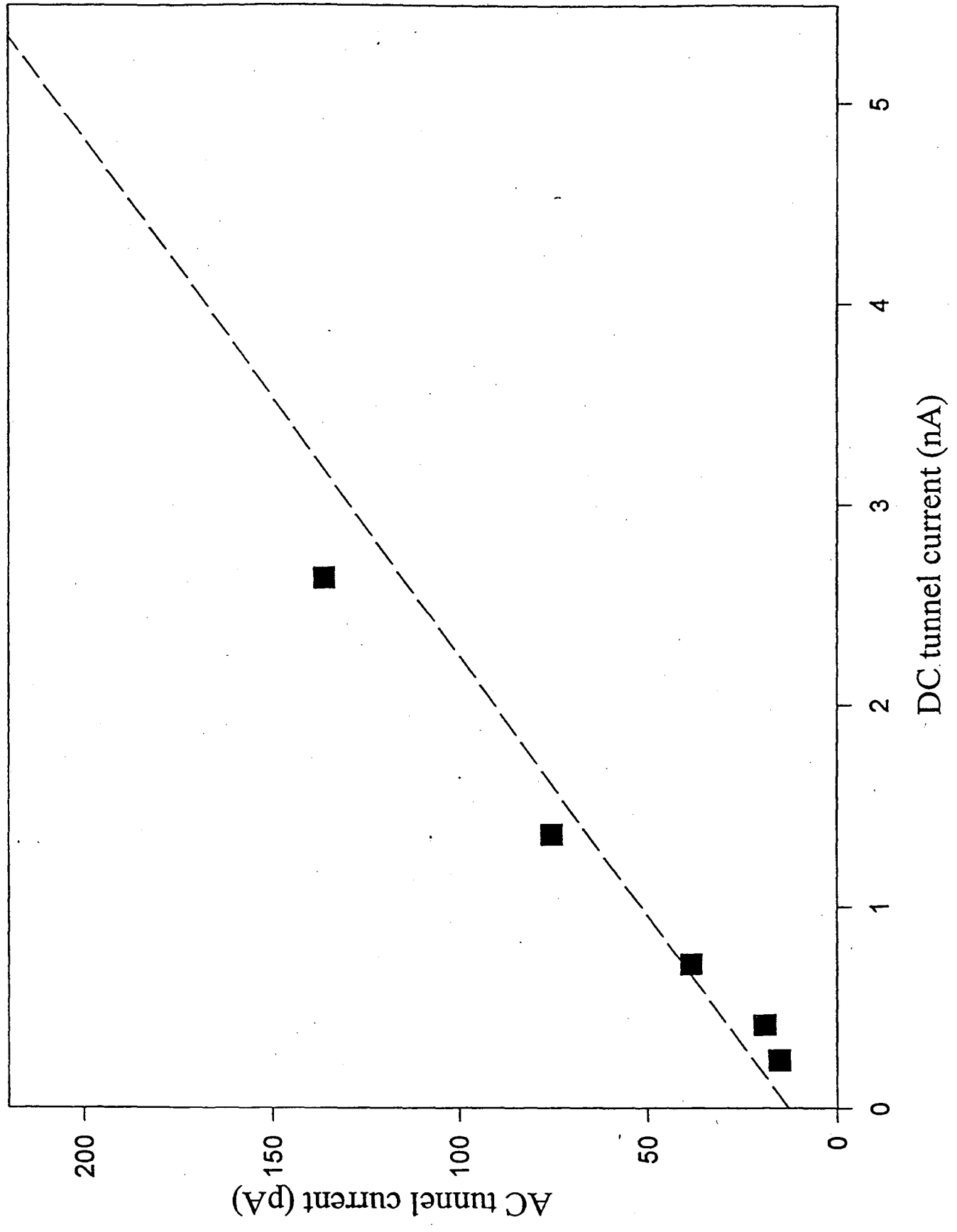


Fig. 3. Plot of AC tunnel current vs. DC tunnel current.

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