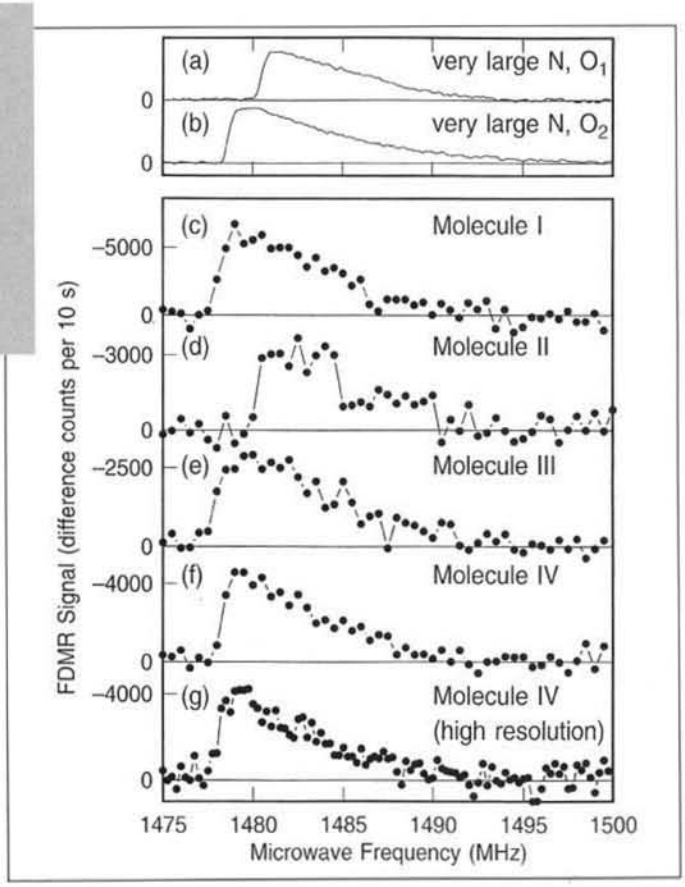
Tx—Tz transition of pentacene in *p*-terphenyl detected with Fluorescence Detected Magnetic Resonance under various experimental conditions. **(a), (b)** Conventional FDMR spectra at large N with excitation in the O_1 and O_2 sites, respectively. **(c)** to **(g)** Single-molecule FDMR spectra for a thin sublimed crystal with different excitation wavelengths of the laser λ = 592.447 nm **(c)**, λ= 592.370 nm **(d)**, λ = 592.404 nm **(e)**, λ = 592.065 nm **(f,g)**]. Traces **(c)** and **(e)** are averages of three; traces **(f)** of eight and trace **(g)** of four individual scans, respectively. Spectra **(a)** and **(b)** are given in arbitrary units. (See Fig. 3 of Ref. 5)



time average. These observations open the way for a variety of new studies of magnetic interactions in solids at the level of a single molecular spin. In particular, the properties of various amorphous organic materials may be able to be studied in greater detail, as the selection of a single molecular spin removes all orientational anisotropy as well as all inhomogeneous broadening. The power of magnetic resonance in general in the study of fine and hyperfine interactions, local structure, and molecular bonding can now be enhanced with these first demonstrations of useful sensitivity in the single-spin regime.

REFERENCES

1. R.H. Clarke, ed. *Triplet State ODMR Spectroscopy*, John Wiley, New York, N.Y., 1982.
2. W. E. Moerner and L. Kador, "Optical detection and spectroscopy of single molecules in solids," Phys. Rev. Lett. **62**, 1989, 2535-2538.
3. M. Orrit and J. Bernard, "Single pentacene molecules detected by fluorescence excitation in a *p*-terphenyl crystal," Phys. Rev. Lett. **65**, 1990, 2716-2719.
4. W. E. Moerner and Th. Basché, "Optical spectroscopy of single impurity molecules in solids," Angew. Chem. **105**, 1993, 537-557; Angew. Chem. Int. Ed. Engl. **32**, 1993, 457-476.
5. J. Köhler *et al.*, "Magnetic resonance or a single molecular spin," Nature **363**, 1993, 242-244.
6. J. Wrachtrup *et al.*, "Optical detection of magnetic resonance in a single molecule," Nature **363**, 1993, 244-245.

Toward Ultrafast Movies of Moving Atoms

BY S. WEISS, D. BOTKIN, D.F. OGLETREE, M. SALMERON, AND D.S. CHEMLA, UNIVERSITY OF CALIFORNIA AT BERKELEY, AND MATERIALS SCIENCES DIVISION, LAWRENCE BERKELEY LABORATORY, BERKELEY, CALIF.

By combining ultrashort laser pulses techniques with scanning tunneling microscopy (STM), we have developed an instrument that obtains simultaneous 2 psec 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 subpicosecond electrical pulse. Our technique is not limited to STM, and can be implemented in a variety of scanning probe microscopies, allowing the observation of ultrafast dynamics on the atomic scale.

The basic idea behind this demonstration is the use of a nonlinear tip-sample interaction to obtain the ultrafast time resolution.[1] This is achieved by modulating both the sample and the tip responses with two short optical pulses. By scanning the time delay between the two pulses and integrating the signal, a cross-correlation of the tip-sample response is obtained. To demonstrate the technique, we used an external nonlinearity, an ultrafast photoconductive switch, photolithographically introduced into the STM tip assembly, rather than the intrinsic nonlinearity of the tunneling gap.

In the experiment (see Figure 1(a)),[2,3] 100 fsec pulses from a modelocked Ti:sapphire laser excited ~650 fsec 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 average tunnel current was recorded as a function of time delay between the two laser pulses. In Figure 1(b), we show a series of time-resolved tunnel current correlations, taken at different tip-sample separations. For clarity, only the changes relative to the average tunnel current are shown. The height of the correlation peak in each trace is approximately proportional to the average tunnel current. Moreover, when the

tip was withdrawn from the surface by 50 Å, both the average and the cross-correlation signals dropped to zero. This means that the observed cross-correlation signal has no contribution from stray capacitance in the leads or from radiative coupling.

Figure 1(c) shows a 0.7×0.7 μm² image of one of the transmission line conductors acquired as the fast pulse passed under the tip. 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 that propagate at velocities of a few Angstrom per femtosecond. 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

The authors would like to thank C. Karadi and P. McEuen for stimulating scientific discussions and invaluable advice, and J. Beeman for the tremendous help with tip fabrication. This work was supported by the Director's Exploratory Research and Development Funds of Lawrence Berkeley Laboratory under the U.S. Department of Energy, contract DE-AC03-76SF00098, and by ONR/ARPA under contract #N00014.93.105.36

REFERENCES

1. S. Weiss *et al.*, OSA Proceedings on Ultrafast Electronics & Optoelectronics **14**, 1993, 162-165.
2. S. Weiss *et al.*, LEOS Optical Microwave Interactions topical meeting digest, Santa Barbara, Calif. 1993, 57-58.
3. S. Weiss *et al.*, "Ultrafast scanning probe microscopy," to be published in Appl. Phys. Lett., Nov. 1, 1993.

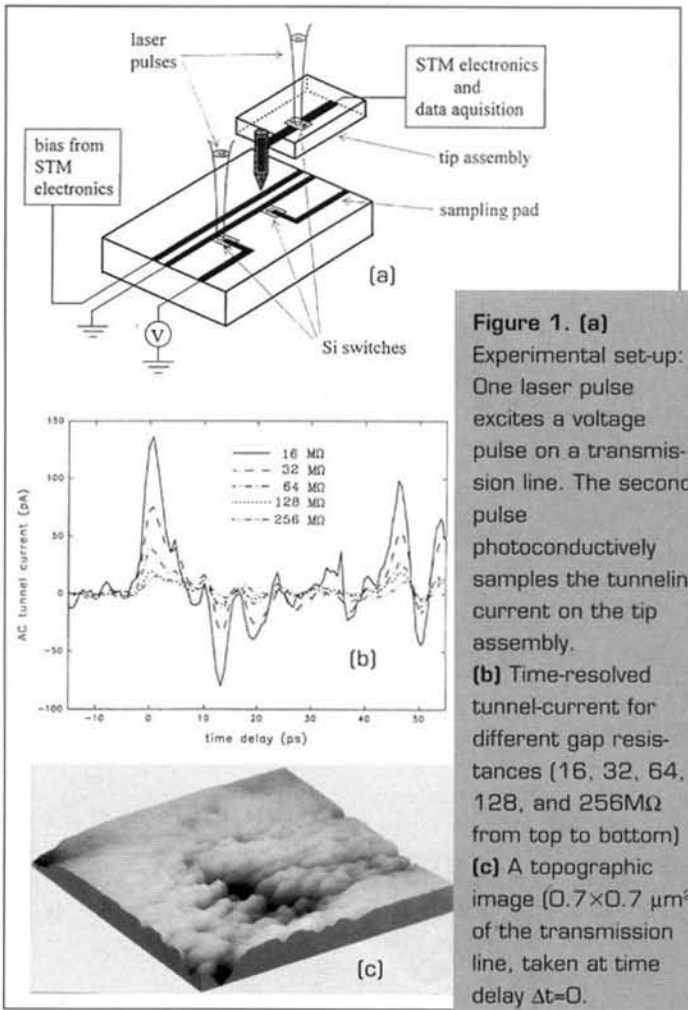


Figure 1. (a) Experimental set-up: One laser pulse excites a voltage pulse on a transmission line. The second pulse photoconductively samples the tunneling current on the tip assembly. **(b)** Time-resolved tunnel-current for different gap resistances (16, 32, 64, 128, and 256MΩ from top to bottom) **(c)** A topographic image (0.7×0.7 μm²) of the transmission line, taken at time delay Δt=0.

Intrinsic Thermal Phase Noise Limit in Optical Fiber Interferometers

BY KEITH H. WANSER, CALIFORNIA STATE UNIVERSITY/FULLERTON, FULLERTON, CALIF.; AND ALAN D. KERSEY AND ANTHONY DANDRIDGE, OPTICAL SCIENCES DIVISION, NAVAL RESEARCH LABORATORY, WASHINGTON, D.C.

Optical fiber interferometers are used for several high sensitivity measurement applications, such as acoustic sensors, magnetometers, accelerometers, and gyroscopes.[1] A variety of noise sources limit the phase shift detection sensitivity attainable using fiber interferometric sensors. The most often quoted (and desired) intrinsic minimum detectable phase shift in interferometric optical systems is the shot, or quantum noise limit. In interferometric optical fiber sensors, the shot noise limit often can be realized; minimum detectable phase shifts of a few μrad/$\sqrt{\text{Hz}}$ at frequencies above a few hundred Hz are readily achieved with modest detectable power levels of a few microwatts.[1] When shot noise dominates, minimum detectable phase shift performance improvements can be obtained simply by increasing the detected power levels.

With the availability of more powerful and extremely low phase noise laser sources such as diode-pumped YAG, considerably lower shot-noise-limited minimum detectable phase shifts should be achievable. The shot noise limit is ~1.5×10^{-8} rad rms/$\sqrt{\text{Hz}}$ for 1.8 mW detected power at 1319 nm. However, workers in the field have not been able to achieve minimum detectable phase shifts below ~$2\text{–}7\times10^{-7}$ rad rms/$\sqrt{\text{Hz}}$ in the important 1-25 kHz band for fiber optic Mach-Zehnder interferometers, regardless of detected optical power. In addition, researchers did not understand why the 1-100 kHz phase noise spectra of fiber optic interferometers exhibits an extremely slow roll off with frequency. This phenomena could not be attributed to laser, electronics, or environmental noise.