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**Keil, Ulrich Dieter Felix; Ha, Taekjip; Jensen, Jacob Riis; Hvam, Jørn Märcher**

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# Femtosecond tunneling response of surface plasmon polaritons

Ulrich D. Keil,[a)] Taekjip Ha,[b)] Jacob R. Jensen, and Jørn M. Hvam
*Mikroelektronik Centret, DK-2800 Lyngby, Denmark*

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We obtain femtosecond (200 fs) time resolution using a scanning tunneling microscope on surface plasmon polaritons (SPPs) generated by two 100 fs laser beams in total internal reflection geometry. The tunneling gap dependence of the signal clearly indicates the tunneling origin of the signal and suggests that nanometer spatial resolution can be obtained together with femtosecond temporal resolution. This fast response, in contrast to the picosecond decay time of SPPs revealed by differential reflectivity measurements, can be attributed to a coherent superposition of SPPs rectified at the tunneling junction. © *1998 American Institute of Physics.* [S0003-6951(98)02223-2]

Time resolved measurements with an ultrafast scanning tunneling microscope (STM) have been made by two approaches: by gating the tunneling signal with a photoconductive switch on the probe[1,2] and by junction mixing.[3] Measurements have so far been performed on transmission lines. Measurements of voltage pulses with a photoconductively gated STM are determined by capacitive coupling and the spatial resolution is therefore limited to tens of nanometers. Generation of the measured signal at the tunneling gap (as in junction mixing) enables the observation of ultrafast effects with the spatial resolution given by the tunneling region. Here, we investigate the rectification of plasmon fields at the tunneling junction and obtain an ultrafast (200 fs) time resolved signal from a scanning tunneling microscope. In this nongated tip configuration of the ultrafast STM, potentially atomic resolution can be achieved along with femtosecond time resolution.

The generation and measurement of surface plasmon polaritons (SPPs) in attenuated total reflection (ATR) offers a sensitive measure of properties and processes at metal–dielectric interfaces. Applications are widespread; the ATR method is also used in biosensing for monitoring biochemical processes at the interface.[4] With a STM, laser induced SPPs have been measured through additional contributions to the tunneling current. These contributions have been explained by heating of the tunneling junction[5] and by rectification of the plasmon field.[6,7] Time resolved reflectivity of SPPs excited with a femtosecond laser source have been reported for different metal surfaces.[8,9] We report on the combination of these techniques.

The sample consists of a 40 nm thick Au layer evaporated on a prism. The SPPs are excited in an ATR–Kretschmann geometry.[10] The experimental setup is shown in Fig. 1. Similar to the measurements in Refs. 8 and 11, we use two laser beams (pump and probe), both *p* polarized and at an incident angle (~42°), to generate the SPPs. The SPP resonance is marked by a drop in the intensity of the reflected light when tuning the incident angle. In the tunneling current measurement, the tunneling tip is placed under the probe beam that is separated by 20 $\mu$m from the pump beam and is in the propagation path of the SPPs generated by the pump beam (Fig. 1). In our setup, we modulate pump and probe beams at fundamental frequencies $(f_0, f_1)$ of at least 600 kHz and measure the signals, either tunneling current or probe beam reflectivity, at the difference frequency of $\Delta f = 1.4$ kHz while the time delay between two beams is varied. (Delay time 0 ps is arbitrary in all measurements.) We find that for these high modulation frequencies, thermally induced signals almost disappear completely (a remaining offset is still seen in the measurements).[12] At low modulation frequencies, we observe heating effects as a dominating offset without delay time dependence. This offset is affected directly by laser fluctuations and leads to a deteriorated signal to noise ratio.

Figure 2 shows the time resolved reflectivity of the probe beam with the characteristic decay time of SPPs on gold. The differential reflectivity is a noncoherent measure of the plasmon decay. The excitation and decay of SPPs leads to a transient increase in the electron temperature which changes the dielectric function of the metal layer.[8] The change of the metal dielectric function implies a shift in the plasmon resonance (versus angle) and is therefore detected as a change in the reflected light at the detector position. We

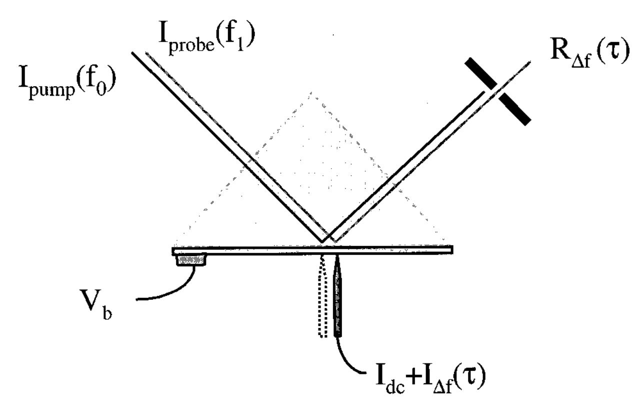


FIG. 1. Experimental setup for time resolved reflectivity and tunneling current measurements. Pump and probe beams are modulated at $f_0$ and $f_1$, respectively, with a difference frequency of $\Delta f$. The change in reflectivity (tip position indicated by dotted outline) and the change in the tunneling current (full tip outline) are measured at $\Delta f$. The laser wavelength is 840 nm and the pulse width 100 fs.

[a)]Electronic mail: ulli@mic.dtu.dk
[b)]Permanent address: Materials Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley, CA 94720.

 

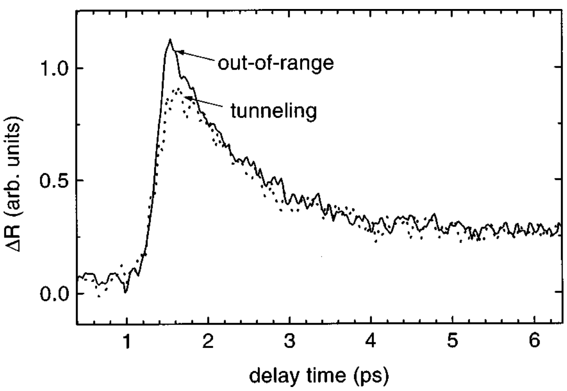


FIG. 2. Differential reflectivity measurement with the tip under the pump beam in tunneling distance and with the tip out-of-range.

observe a difference between measurements with the tip in tunneling range and with the tip withdrawn (~1 μm). The tip is positioned under the pump beam. With the tip in tunneling range, the signal is attenuated during the first 300 fs. This is evidence that coherent effects contributed to the initial short time response. Because the plasmon field extends into the air on a near micrometer scale, the tip acts as an efficient scatterer for plasmons[13] that destroys their coherence if it is in range. We note that this observation is consistent with measurements in Ref. 11 where a reflectivity change is measured for a modulated tip-distance (albeit with overlapping beams). In Ref. 13 the scattering of plasmons with a tunneling tip is investigated and used to influence the plasmon decay with a lateral resolution of 3 nm. Figure 3(a) shows the delay time dependent change in the tunneling current. The signal width of 200 fs full width half maximum (FWHM) is at the resolution limit of the setup. The signal disappears completely if the polarization direction of one or both beams is rotated by 90° indicating that the time resolved signal arises from SPPs. The dependence of the signal amplitude on the dc tunneling current is shown in Fig. 3(b). As the measurements are performed for a constant bias ($V_b=48$ mV) in constant current mode, a higher current implies a shorter tip-sample distance. The fit included in the graph shows that the dependence on the tunneling current follows approximately a square-root dependence. If the tip is retracted out of the tunneling regime, the signal disappears completely. For overlapping beams,[11] a signal due to multi-photon ionization had been observed for tip-sample distances up to 1 mm. As we observe a fundamentally different for separated beams, we rule this contribution out and concentrate in the discussion on rectification effects.

Several authors[5,6] report on the detection of SPPs with a STM through thermal effects and a rectified signal. The rectification contribution, however, was only resolved at certain locations with a pronounced nonlinearity of the tunneling current–voltage ($I-V$) curve. We do not observe such locations. On the other hand, only rectification effects are fast enough to be resolved in a measurement on a femtosecond timescale. As our measurement suppresses slow, thermal effects, it is more sensitive to smaller, otherwise hidden, rectification effects.

For a careful comparison of the relation between the $I-V$ curve nonlinearity and the plasmon induced signal amplitude, we utilize a sensitive measurement of the nonlinearity at low (near dc) frequencies. From expanding the $I-V$ curve around a given bias point $V_b$, we obtain:

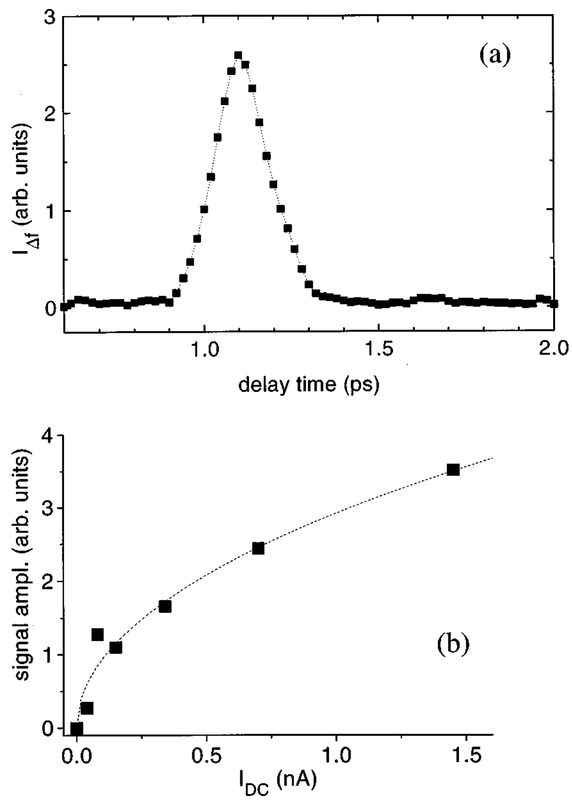


FIG. 3. (a) Delay time dependent tunneling current measured at $V_h=48$ mV, $I_{DC}=0.7$ nA with the tip under the probe beam. (b) Amplitude dependence on $I_{DC}$. The data are compared to a fit function $y=a\,x^b$ with $b=0.49$.

$$I(V_b+V_0\sin\,\omega t)=I(V_b)+\left.\frac{\partial I}{\partial V}\right|_{V_b} V_0\sin\,\omega t + \left.\frac{\partial^2 I}{\partial V^2}\right|_{V_b} \frac{V_0^2}{4}(1-\cos\,2\,\omega t). \quad (1)$$

This expansion shows that the second derivative determines an additional dc contribution to the current (the rectified signal) but it also shows that this second derivative can be measured at the second harmonic of the modulation frequency. This is equivalent to optical frequencies where the rectified signal and second harmonic generation are determined by $\chi_2$.[14] We perform this measurement by modulating the bias voltage with $\omega=2.5$ kHz and detecting the second harmonic in the tunneling current with a lock-in amplifier (Fig. 4). According to Eq. (1), this measurement gives us a measure of the expected rectified signal at kHz (or near dc) frequencies. The modulation frequency is chosen to be above the cutoff frequency of the feedback loop and below the cutoff of the current amplifier. We point out that the $I_{DC}$ dependence of the femtosecond cross-correlation signal and the kHz modulation signal are both performed with the feedback loop on and that the $I_{DC}$ dependence is also a measure of the distance dependence.

For optical frequencies the term $V_0\sin\omega t$ represents the additional voltage induced by the plasmon field. The two laser beams generate the plasmon voltages $V_0\sin\omega t$ and

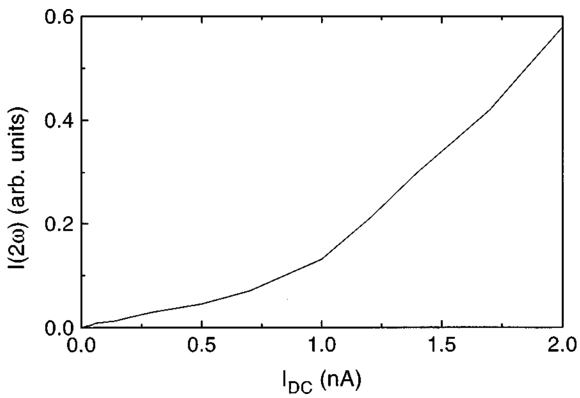


FIG. 4. Second harmonic tunneling current contribution, $I(2\omega)$, for a bias modulation at $\omega$=2.4 kHz. $I_{DC}$ is set average tunneling current, the bias voltage is $V_b=48$ mV, and the voltage modulation amplitude is $V(\omega)=10$ mV.

$V_1 \sin(\omega t+\delta)$. The equation illustrates that we can only expect a delay time dependent signal for a coherent superposition of the plasmon fields generated by the two laser beams. This means that, although the SPPs decay on a time scale of a picosecond, a signal is only observed during the coherence time of the plasmon wave. For times longer than the laser pulse, the SPPs lose their fixed phase relation due to scattering. The comparison of the amplitude dependences in Fig. 3(b) (optical frequencies) and Fig. 4 (kHz frequencies) shows a slower increase of the rectified signal in Fig. 3(b) with increasing $I_{DC}$. In other words, for decreasing tip-sample distance the $I-V$ nonlinearity increases more strongly than the time resolved plasmon signal. This deviation is consistent with the fact that the tip itself destroys the coherent signal as indicated by the reflectivity measurement. The closer the tip is, the more the tip influences the plasmon field. This effect can compensate for the higher second order nonlinearity observed for high $I_{DC}$ at low frequencies.

In summary, we presented the time resolved detection of surface plasmon polaritons with a scanning tunneling microscope. The results indicate that the time resolved signal is due to rectification of coherently superimposed plasmon voltages. The fact that the signal disappears outside of the tunneling range should imply a comparable lateral resolution which needs to be tested in future investigations. The comparison with differential reflectivity measurements shows that the tip itself influences the decay of the plasmon field coherence.

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