Laser field enhancement at the scanning tunneling microscope junction measured by optical rectification

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In this letter we report the measurement of the field enhancement at the tip of a scanning tunneling microscope, by means of the detection of the optical rectification current. A field enhancement factor between 1000 and 2000 is obtained for highly oriented pyrolytic graphite and between 300 and 600 for gold. Field enhancement factors found are strongly dependent on the particular tip used. The magnitude of the emitted light at the field enhanced region, calculated from the measured optical voltage, could be easily detected by a simple photodiode. © *1998 American Institute of Physics.* [S0003-6951(98)01317-5]

The near field scanning optical microscopy (known as SNOM or NSOM)[1] has provided an increase of the resolution of optical microscopes by several orders of magnitude. These microscopies rely in the use of an aperture to chop the wave front and the achievable resolution is of the order of tens of nm, limited by fact that the aperture cannot decrease beyond the skin depth of the light.[2] Alternative apertureless schemes have proved to be the solution to this limitation, relying in the emission from the light-induced dipole at the dielectric tip of a force microscope.[3,4] An alternative scheme would be to use the field enhancement at a metallic tip, arising from the plasmon resonance, as found in surface enhanced raman spectroscopy.[5] The field enhancement provides an excellent source for linear and nonlinear microscopies with high spatial resolution. The actual value for the field enhancement has never been measured, and theoretical calculations predict enhancement factors larger than 1000, strongly dependent on the tip–sample distance, geometry of the tip and sample, and dielectric response of the tip and sample materials at the optical frequency.[6,7] In this letter, we report the measurement of the field enhancement at the tip of a scanning tunneling microscope (STM), by means of the detection of the optical rectification current. Field enhancement factors larger than 1000 are found, strongly dependent on the particular tip used.

Figure 1 shows the experimental setup used. A low power laser is modulated before being focused into the tip–sample junction. The spot size was about 100 μm×50 μm and the incidence angle was 60° with the horizontal. Results were obtained in samples of highly oriented pyrolytic graphite (HOPG) and a gold foil of about 0.3 mm thickness. The STM tip is approached without light illumination until the desired tunneling current is obtained at a set tip–sample voltage. The current is preamplified and measured by the STM control module. Time constant of the feedback loop is set to correct drifts of low frequency but does not respond to the frequencies used for the light modulation. Usual $I-V$ curves are measured applying a voltage ramp to the tip at constant tip–sample distance. The modulated component is measured by a lock-in amplifier in series with the current loop, with a reference signal coming from a photodiode that samples the laser light. The STM current–voltage converter has a known frequency dependence with a cutoff at about 8 kHz; therefore, all the modulated signals have been corrected by this response. The lockin output may be reinjected in the STM data acquisition module in order to record $I-V$ curves of photoinduced current simultaneously with normal ones. In our system the sign of the dc voltage coincides with the polarity of the tip. The light is polarized with the electric field in the incidence plane (p polarization), so that a net component appears in the tip–sample direction.

If the electric field from the light is enhanced at the tip, it should induce a voltage between the tip and sample at the optical frequency ω_l, $Vi=\nu\cos(\omega_l t)$. If the tunneling is fast enough, this modulated field should give rise to a rectified contribution arising from the series expansion:

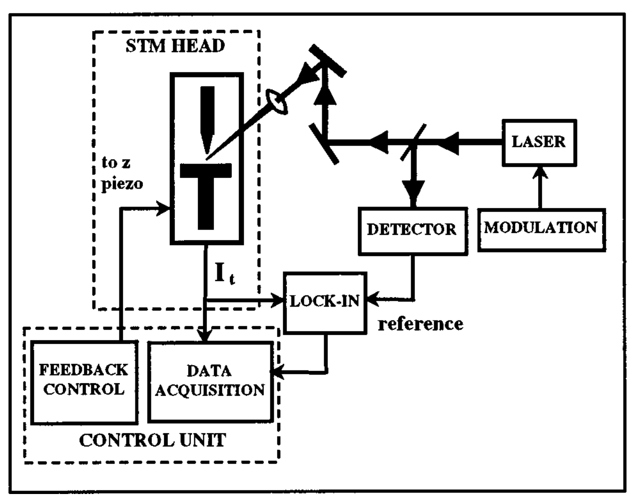

FIG. 1. Experimental setup. An electrical modulated low power p polarized laser diode ($\lambda=670$ nm) is focused into the STM junction by a 70 mm focal length objective. The tunneling current and the modulated current are measured, and the tunneling current–voltage ($I-V$) curve and photoinduced current–voltage (I_p-V) curve can be recorded simultaneously. The detector samples the laser emission, in order to obtain an absolute reference for the phase.

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$$I = I(V_b) + \left(\frac{\partial I}{\partial V}\right)_{V_b} V_i + \frac{1}{2}\left(\frac{\partial^2 I}{\partial V^2}\right)_{V_b} V_i^2 + \cdots , \quad (1)$$

where V_b is the bias voltage. The linear term averages to zero, and the second derivative term is proportional to the laser intensity and hence will appear in the lock-in in phase with the light modulation. This term was shown to appear by Völker *et al.*[8,9] for the infrared at GHz modulation frequencies.

As the light is being modulated, a thermal expansion contribution to the tunneling current appears at the modulation frequency ω. In a previous work we have shown[10] that the thermal expansion contribution will roll off as $1/\omega$, and the in phase contribution as $1/\omega^2$, above a cutoff frequency that depends on the thermal diffusivity of the sample and the laser beam spot size. Unfortunately, due to the limited bandwidth of the current amplifier of the STM, driving the system above the cutoff frequency would require very large spot sizes, that would reduce the beam intensity below detectable levels of light-induced currents. The thermal contribution cannot be avoided, but can be discriminated by using the fact that the thermal expansion gives rise to a change in the gap distance, that for small biasing voltages (less than 0.5 V), yields a modulated current I_T following an $I_T - V$ curve proportional to the static $I-V$ curve.[10] Hence the measured modulated current (I_p) will have two contributions, one of thermal origin proportional to the background current, and one from the optical rectification following the last term of Eq. (1):

$$I_p = aI + b\,\frac{\partial^2 I}{\partial V^2} + c. \quad (2)$$

The coefficient a accounts for the in-phase thermal expansion contribution. The term with b accounts for the optical rectification and the constant term was added in order to account for the observed offset, and will be discussed later.

$I-V$ and $I_p - V$ curves were recorded for different samples (HOPG and Au), for different tips and at different set points of the STM (that would correspond to different tip–sample distances). Two typical cases are shown in Fig. 2 for gold and Fig. 3 for HOPG, obtained both with the same tip (mechanically cut Pt–Ir tip) and the same laser power and focusing conditions. The modulated current versus voltage curves were fitted by Eq. (2) for both in-phase and quadrature components, and the partial regression coefficients were obtained for the least square fit. The standard partial regression coefficients β, expresses the relative standardized strengths of the effects of the different independent variables on the same dependent variable in a multiple regression.[11] In all cases a negligible contribution from the optical rectification was obtained from the fit for the in-quadrature component (in the inset of the Figs. 2 and 3) as well as for the curves recorded with s polarized light (no component of the electric field in the tip–sample direction), while the in-phase component always requires a larger relative weight of the optical rectification. The constant term was not present if the light was blocked, indicating it is of optical origin.

From the fitted value of parameter b and using Eqs. (1) and (2), the light-induced voltage drop across the tip–sample junction can be computed (due to the small size, the electrostatic potential can be used), yielding 12 ± 0.6 mV for Au and 38 ± 3 mV for HOPG. Assuming a tip–sample distance between 0.5 and 1 nm, which are conservative values for typical STM conditions, and from the laser power (10 mW) and beam size at the sample (100 μm$\times$50 μm), a field enhancement factor between 1000 and 2000 is obtained for HOPG and between 300 and 600 for gold. The same measurement repeated with other tips yielded somewhat smaller values.

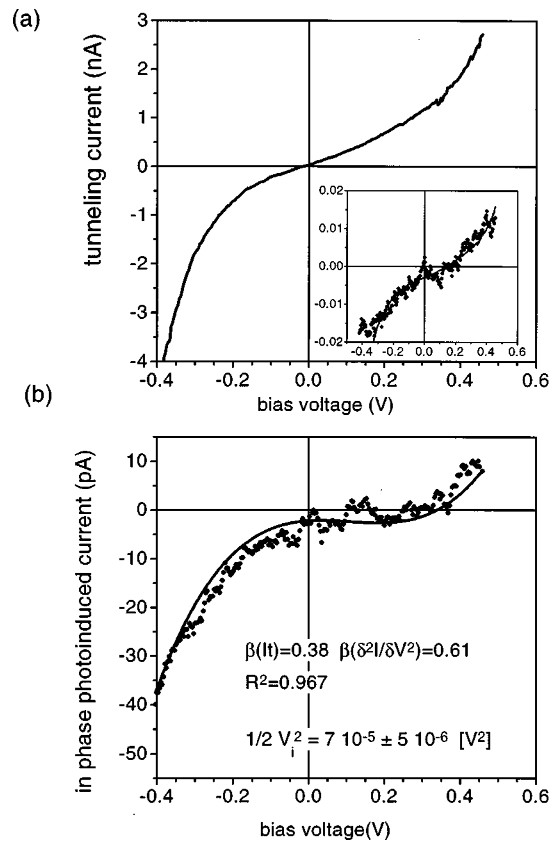

FIG. 2. (a) Tunneling current vs bias voltage curve for a foil gold sample and Pt–Ir tip. The setpoint was set at $V_t = -100$ mV, $I_t = 0.2$ nA. The inset shows the 90° dephased component of the photoinduced current, where the full line is the fit with the tunneling current, making evident the thermal nature of this component. (b) In-phase photoinduced current vs bias voltage. The β^2's are the standard partial regression coefficients. R^2 is the regression coefficient. The full line is a fit including a linear dependence with the second derivative of the static curve [see Eq. (2)]. The value of the rectified voltage (show in the figure) can be calculated from the fitted parameters. Laser intensity$=3\times10^5$ W/m^2. Modulation frequency$=$10 kHz.

The constant terms added to the fit were always of the order of magnitude of the noise (a few pA), so that no systematic measurement was possible to clearly pin down its origin. Modulation of the capacity between the tip and the sample due to the thermal expansion yields values too low to account for the observed offset. Direct optical transitions between the tip and the sample is one possible mechanism to explain the offset, once the field enhancement is taken into account. In fact, the intensity of 3×10^5 W/m^2 enhanced by 10^6 yields a cross section of 10^{-19} cm^2 if the induced current is 3 pA. This number is within typical values for atomic dipole transitions, and considering that the local nature of the field and the fact that the transition is between neighboring atoms, no selection rules apply, many states contribute to the transition and the dipole moment should be large. This

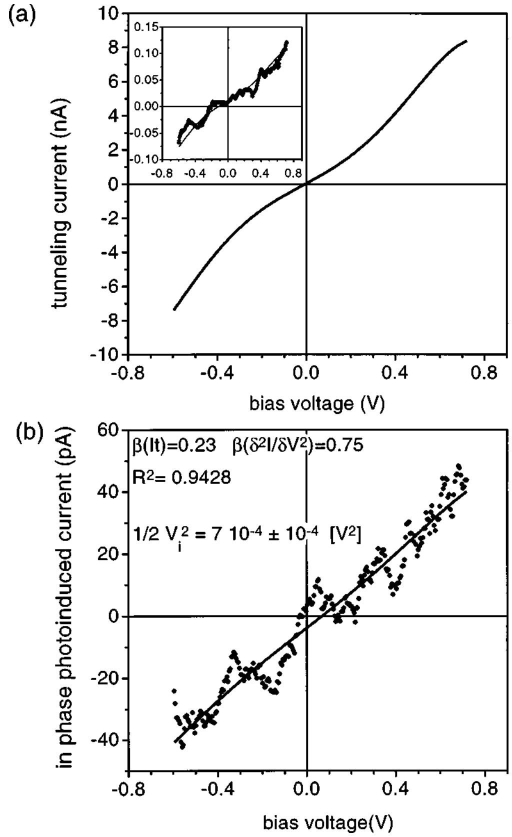

FIG. 3. (a) Tunneling current vs bias voltage curve for a HOPG sample and Pt–Ir tip. The setpoint was $V_t = -100$ mV, $I_t = 0.5$ nA. The inset shows the 90° dephased component behavior. The full line is the fit with the tunneling current. (b) In-phase photoinduced current vs bias voltage. The β^2's are shown in the figure. The full line is a fit including a linear dependence with the second derivative of the static $I-V$ curve. The value of the rectified voltage (show in the figure) can be calculated from the fitted parameters. Laser intensity$=3\times10^5$ W/m^2. Modulation frequency$=4$ kHz.

mechanism can only account for the in-phase part of the observed offset. Another optically induced current source that could have time delays (and have in-quadrature contributions), is that arising from optically excited charges at the surface contamination layer always present in ambient STM (water and adsorbates). The order of magnitude of the observed offset, would correspond to one electron per 10^9 photons, that represents a very small transition yield. In any case, the observed offset is too small for a more detailed study with our present instrumentation, and does not represent a relevant contribution to the total induced current except at $V=0$.

The large enhancement of the field obtained should provide an easily detected emitted power from the field enhanced region. To estimate the order of magnitude of the emitted power expected, a very simple naive model was used. The light was assumed to induce a dipole at the tip (of radius R), and an image at the sample. Requesting a constant potential surface at the tip and sample, and a voltage drop V_i between the tip and the sample (located at a distance d), the position and moment (p) of the tip dipole can be computed, yielding for $R \gg d$:

$$p = 9V_i(R^3 d/6)^{1/2} \tag{3}$$

that would radiate a power between 0.1 and 100 nW for $d = 0.5$ nm and R between 10 nm and 100 nm. These values are in agreement with the 2 nW collected power reported by Caldarone *et al.*[12] for a gold covered silicon sample, in very similar experimental conditions.

In conclusion, the optical field enhancement at the STM junction has been measured for platinum tip and HOPG and gold samples, yielding for some cases values larger than 1000, depending on the particular tip used. These values indicate that the field enhancement should provide a unique tool for a new type of microscopy (field enhanced scanning optical microscopy) and moreover for nonlinear optical microscopy and spectroscopy.

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