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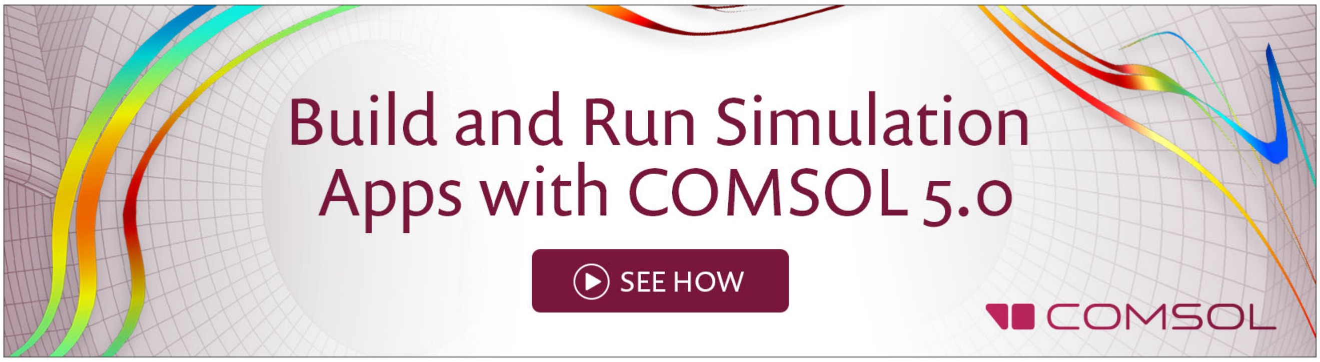

APPLIED PHYSICS LETTERS **101**, 213104 (2012)

# Tip expansion in a laser assisted scanning tunneling microscope

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The thermal expansion of a scanning tunneling microscope tip induced by femtosecond laser is investigated with various parameters including laser power, modulation frequency, illumination spot, and laser wavelength. The magnitude of tip expansion is measured to be proportional to the laser power. The response bandwidth is closely related to the length of the tip cone section, which is consistent with a two-rod model simulation. While visible lasers produce significant tip expansion, deep ultraviolet and near infrared lasers result in significantly reduced expansion magnitude, which can be explained with the tip induced surface plasmon in the tunneling junction. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4767877]

The combination of scanning tunneling microscope (STM) with optical excitation, which is described as laser-assisted STM,[1–6] envisioned the possibility of exploring light-sensitive dynamic processes with the highest spatial resolution. Significant progresses have been made in recent years including molecular motion induced by laser pulses,[4] coupling between photon and tunneling electrons,[7] and carrier relaxation with nanometer scale resolution.[8–10] When introducing laser into a STM tunneling junction, the tip expansion due to laser illumination has to be considered carefully, because the tunneling current is extremely sensitive to the tip-sample distance (roughly, current may change by one order of magnitude if the junction width changes by 1 Å) and the sample may be impaired if the tip expands too much and faster than the STM feedback servo. Experimental techniques have been developed to minimize the effect of tip expansion, such as retracting tip during laser illumination,[4] employing continuous wave (CW) laser without modulation,[7] and utilizing shaken-pulse-pair excitation techniques.[8–10]

Despite the success by using these techniques, understanding the dynamic behavior of tip expansion due to laser illumination is still of great concern in the development and application of laser-assisted STM, especially in cases of illumination with pulsed or modulated laser. The effect of CW laser illumination on STM tip expansion has been extensively investigated with various tips and samples, as well as analytical models.[11–13] Transient tip expansion due to femtosecond laser pulses has also been measured with AFM cantilever.[14] To date, direct measurement of tip expansion under illumination of femtosecond laser in a STM setup has not been carried out. In addition, while it is common that light absorption, and thus thermal expansion, is closely related to the incident laser wavelength, the response of a STM tip under illumination of different wavelengths is still unknown.

In this letter, we investigate the thermal expansion of a STM tip illuminated with femtosecond pulse lasers of various wavelengths, ranging from deep ultraviolet (DUV) to near infrared (NIR). The tip response is measured according to various parameters including laser power, modulation frequency, illumination spot, and laser wavelength. The dynamic behavior of the tip expansion is compared with a few model simulations for better understanding. The frequency response of the tip expansion has been measured as a function of the position of illumination. The influence of laser wavelength is discussed based on tip induced surface plasmon that determines the photon absorption efficiency.

Figure 1 shows the experimental set-up, which consists of a home-built ultrahigh-vacuum (UHV) STM[15] and a femtosecond laser system. The laser system is built with a Ti-sapphire source laser (Coherent Chameleon Vision II) that delivers 150 fs pulses at 80 MHz repetition rate, with wavelength tunable between 680 and 1080 nm. By using two stages of second harmonic generators (SHG), lasers with wavelength at 340–540 nm and 175–185 nm are also available. One of the advantages of this system is, thus, that the laser wavelength can be easily tuned to cover several bands ranging from DUV to NIR. The laser beam is directed onto the STM tunneling junction at an incident angle of 45°, with a beam diameter of about 50 $\mu$m. The STM tip-sample junction and the illumination spot can be monitored by a CCD mounted on the other side of the UHV chamber. The inset of Figure 1 shows a typical CCD image as obtained, where the tip, mirror image of the tip, and the laser spot are present. In our experiment, the STM is operated in the constant-current mode at room temperature, and an electrochemically etched tungsten tip and a polycrystalline copper sample are employed. The laser beam is turned on and off by a mechanical shutter, and the STM tip expansion is measured by recording feedback control voltage on the Z piezo tube at a rate of 5000 samples per second. To investigate the frequency response, the laser beam is modulated by a chopper at a frequency range of 10 Hz–1 KHz, and the corresponding

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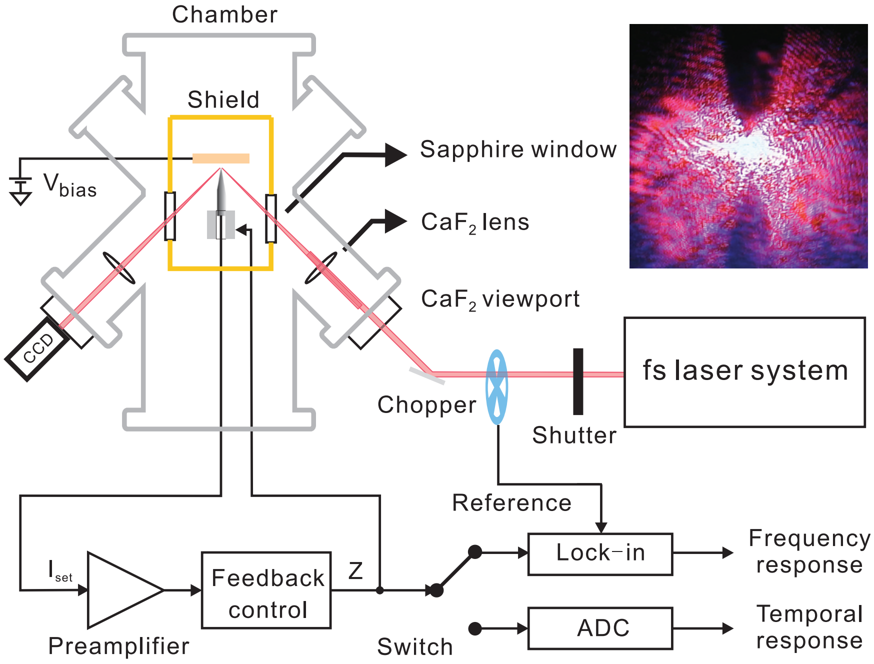

FIG. 1. Experiment set-up. The system consists of an UHV STM and a femtosecond laser system. The tip expansion is measured by monitoring feedback control voltage on the Z piezo tube, either through a lock-in amplifier for frequency response or an analog-digital converter (ADC) for temporal response. The inset shows a typical CCD image of the STM junction under laser illumination.

modulation in tip length is measured with a lock-in amplifier (Signal recovery, model 7625).[11–13]

The dynamic behavior of STM tip expansion under laser illumination can first be understood by theoretical models and simulations.[12,13] Normally, only the expansion along tip axis (z direction) is concerned because of its larger vertical scale. In addition, the thermal expansion of the sample is usually negligible compared to the tip.[12] The thermal expansion of the sample in our experiment is estimated to be two orders of magnitude smaller than the total expansion of the tip. When a modulated laser illuminates the tip apex, it generates a thermal wave that propagates along the axis and creates an oscillation of expansion. The thermal wave damps to 1/e of its initial intensity after a distance equals diffusion length, $D = \sqrt{2\kappa/\omega}$ where $\kappa$ is the thermal diffusivity and $\omega$ is the modulation frequency. The magnitude of tip expansion has a strong dependence on modulation frequency. For low frequency modulation such that the diffusion length is larger than the length of the tip $l$, i.e., $\omega < 2\kappa/l^2$, the magnitude of tip expansion is almost independent of modulation frequency. As modulation frequency increases to higher than a cut-off frequency, $\omega_0 = 2\kappa/l^2$, there is a significant decrease in the magnitude of tip expansion as modulation frequency increases.

Figure 2(a) shows the calculated expansion of a uniform tungsten rod as a function of modulation frequency. In the calculation, we employed coefficient of thermal expansion $\sigma = 4.5 \times 10^{-6}\mathrm{K}^{-1}$, tip length $l = 10\,\mathrm{mm}$, heat conductivity $\lambda = 173\mathrm{W}/(\mathrm{m}\cdot\mathrm{K})$, cross-section of rod $S = 1.96 \times 10^{-7}\mathrm{m}^2$, and the laser power $P = 1\,\mathrm{mW}$. The inset shows the temperature distribution along the rod at the cut-off frequency $\omega_0$ of 0.22 Hz. For modulation frequency $\omega < \omega_0$, the magnitude of tip expansion is about 66.7 Å. For $\omega > \omega_0$, the tip expansion is approximately inversely proportional to the modulation frequency.

The simple rod model, however, cannot simulate well the experimentally measured tip behavior under laser illumination. This is because the electrochemically etched tungsten tip has a cone shape end, as shown in the inset of Figure 2(b), which results in significant difference in thermal conductivity. A parabolic end model,[13] a cone end model,[12] and a two-rod model are employed for more realistic simulation. The length of the tip end is set to 0.9 mm, which is the cone length of the tip used in our experiment. Figure 2(b) shows the numerical calculations with all three models. All three models result in a low cut-off frequency around 0.22 Hz and a high cut-off

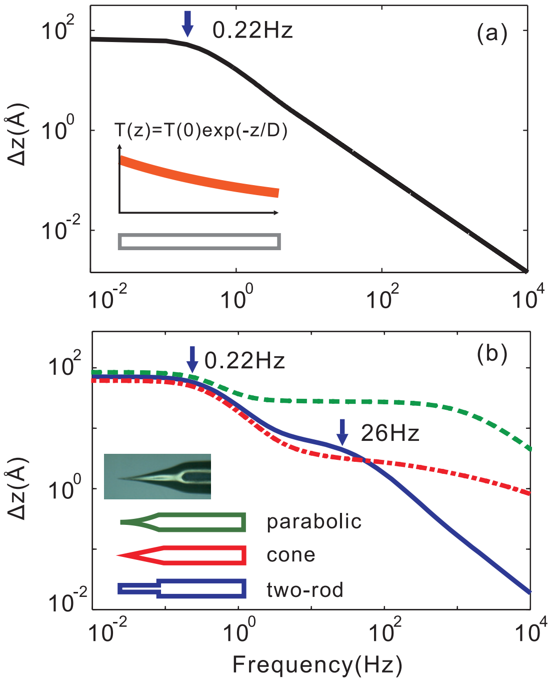

FIG. 2. Simulation of STM tip expansion under laser illumination. (a) Thermal expansion of a simple tungsten rod as a function of laser modulation frequency. Inset: temperature distribution along the rod at the cut-off frequency of 0.22 Hz. (b) Frequency response of three tip models: a two-rod model (blue solid line), a cone end model (red dashed dotted line), and a parabolic end model (green dashed line). The optical microscope image of a tip and three models are shown in the inset. The white fork-like contrast in tip image is due to the top light illumination.

frequency between 20 Hz and 2 kHz. The low cut-off frequency is determined by the total length of the tip, and it is thus not sensitive to the tip shape. For a tip length of 10 mm in our experiment, it can be estimated to be 0.216 Hz by using $\omega_0 = 2\kappa/l^2$, which matches well with the value in the simulation. The significant variation in the high cut-off frequency indicates that the fast dynamic behavior of tip expansion is very sensitive to the tip geometry.

Figure 3(a) shows a typical response of the tip expansion when laser illumination (800 nm, 433 $\mu$W) is turned on to the STM junction. Since the 80 MHz repletion rate is much faster than the STM feedback electronics (10 kHz bandwidth), transient response from each laser pulse is undetectable with this method. We find that the tip expansion can be fitted with an exponentially decay function with a linear term,

$$\Delta Z(t) = z[1 - \exp(-t/t_0)] + kt, \tag{1}$$

where $t_0$ is a time constant, which reflects the slow expansion of the shank, and $z$ represents the magnitude of the expansion. The coefficient $k$ in the linear term represents the continuous heating up of the tip due to laser illumination. It is not feasible to derive the information about the fast expansion in cone region by fitting the temporal expansion curve in our experiment, due to the low bandwidth (50 Hz) in the data acquisition to minimize the noise.

By varying the laser power between 0.2 mW and 0.5 mW, we find that the time constant remains unchanged while the tip expansion $z$ is linearly proportional to the incidence power, as shown in Figure 3(b). The coefficient $k$ is also linearly proportional to the incidence power, as expected.

The dynamic behavior of tip expansion under laser illumination can also be investigated by measuring its frequency response, where the laser beam is modulated with a chopper in a frequency range between 10 Hz and 1 kHz. Fig. 3(c) shows a typical response curve under the illumination of 800 nm laser at 533 $\mu$W. The magnitude of expansion decreases as the modulation frequency increases, and it approaches *1/f* at frequencies higher than the cut-off frequency. A cut-off frequency of 24 Hz can be identified by extrapolation of fitted lines, which is consistent with the simulated cut-off frequency of 26 Hz with the two-rod model. This is very surprising since the cone end model and parabolic end model represent the real tip geometry much better than the two-rod model. The magnitude of tip expansion as a function of laser power is shown in Figure 3(d), revealing a linear dependence as theoretical analysis predicts.

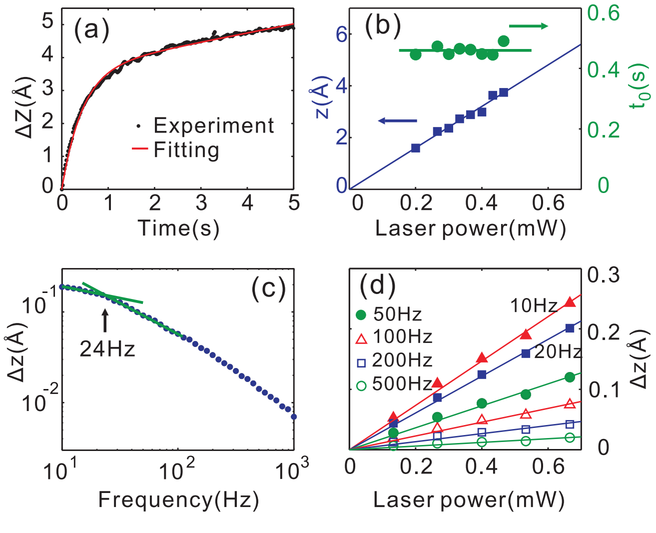

FIG. 3. Dynamic behavior of tip expansion under illumination of femtosecond laser with different power (laser wavelength at 800 nm). (a) Temporal response of tip expansion when laser hits the STM junction, with laser power of 433 $\mu$W. Experimental curve is fitted with an exponentially decay function and a linear term. (b) Derived $z$ and $t_0$ as a function of laser power. (c) Frequency response of tip expansion with modulated laser from 10 Hz to 1 kHz. (d) The magnitude of tip expansion as a function of laser power at different modulation frequencies.

To better understand the influence of laser illumination on tip response, different illumination spots have been checked for changes in tip response. The inset of Figure 4(a) presents a CCD image of STM junction where the lower part is the real tip and the upper one the mirror reflection from the sample surface. The illumination spots are chosen along the tip axis. The tip end is referred to as the origin, and the relative position of each spot is $-327\ \mu$m, $0\ \mu$m, $101\ \mu$m, and $556\ \mu$m along the tip axis, as indicated with the dashed lines. The positive position indicates that the laser focus is on the sample and then reflected to the tip, which is equivalent to the corresponding negative position except the reduction of illumination intensity due to surface reflection.[12] Figure 4(a)

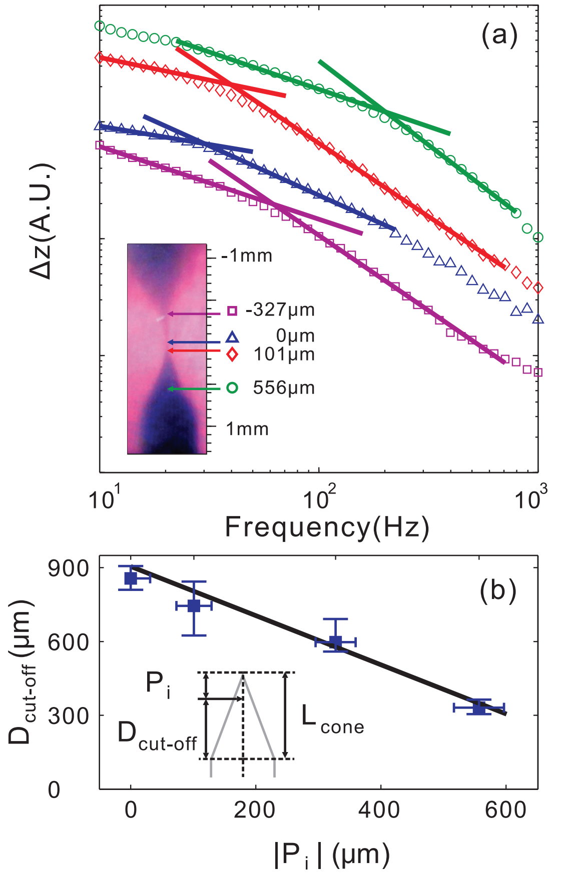

FIG. 4. (a) Frequency response of tip expansion under laser illumination at four different spots. Laser wavelength is 800 nm and the incidence power is between 200 $\mu$W and 400 $\mu$W. Curves are shifted for clarity. Straight lines are guide for obtaining the cut-off frequencies. Inset: CCD image of STM junction with illumination spots indicated with dashed lines. (b) The diffusion length at the cut-off frequency versus illumination position. Inset: schematic of relation between the diffusion length and the illumination spot.

shows the frequency response of tip expansion with different illumination spots. Cut-off frequencies of 46.0 Hz, 26.8 Hz, 31.0 Hz, and 166.5 Hz are derived from the corresponding curves. The diffusion lengths at the cut-off frequencies, $D_{cut-off}$, are then calculated and plotted versus the illumination position $P_i$, as shown in Fig. 4(b). It is interesting to notice that the diffusion length has a linear dependence on the illumination position,

$$D_{cut-off} = L_{cone} - P_i, \tag{2}$$

where $L_{cone} = 905\ \mu\text{m}$ is the total length of the tip cone.

Energy transfer from the illuminating laser to the tip-sample junction is essential in determining the magnitude of tip expansion. To understand such energy transfer mechanism and for practical reasons, it is beneficial to investigate the tip expansion with lasers of different wavelengths. Four laser wavelengths of 800 nm, 488 nm, 360 nm, and 177 nm are chosen in our experiment. Figure 5(a) shows the frequency response of tip expansion with normalized with the incidence power for wavelengths of 800 nm, 488 nm, and 360 nm. The tip expansion signal under illumination of 177 nm laser is too weak to be detected, and thus is not shown in the figure. For better comparison, the normalized magnitude of tip response at 50 Hz modulation frequency versus wavelength is shown in Figure 5(b). It is clear that tip expansion at laser wavelength of 360 nm and 488 nm is dramatically larger than that at 800 nm and 177 nm. Especially, the tip expansion under illumination of 177 nm DUV laser is indistinguishable from the background noise and only the noise level is shown in the figure as the estimated maximum value.

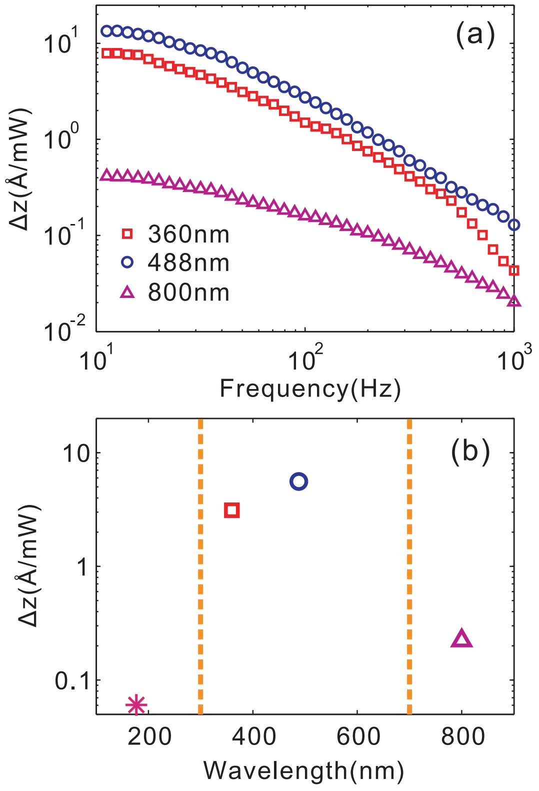

FIG. 5. (a) Frequency response of tip expansion under the laser illumination at wavelength of 800 nm, 488 nm, and 360 nm. (b) Normalized magnitude of tip expansion at modulation frequency of 50 Hz. The dashed lines indicate the range of plasmon emission spectrum of tunneling junction with tungsten tip and Cu(111) sample (Ref. 17).

The significant difference in energy transfer efficiency with lasers of various wavelengths cannot be explained solely with the absorption spectrum of tungsten, which shows a rather flat feature from 300 nm to 800 nm.[16] Instead, this may be related to the photon absorption with local plasmon at the tip-sample junction. Several tip-induced plasmon modes on Cu surface in the range between 200 nm and 800 nm, with an emission peak around 600 nm, are elucidated by light emission characteristics from the tunneling gap, as well as theoretical calculations.[17–19] This is consistent with our experimental measurement, where 488 nm laser produced a maximum magnitude of tip expansion.

We also note that the photo-excited hot electrons will modulate the local density of states (LDOS) and thus the tunneling current, which eventually contributes to the z-piezo change. The experimentally observed LDOS change, however, is pretty weak. Assuming a 10% change in LDOS, the upper limit of hot electron induced z-piezo change is estimated to be 2 pm, much smaller than the physical expansion of the tip. This validates our assumption that the z-piezo change is dominated by the STM tip.

Our experiment demonstrated that dynamic behavior of tip expansion under illumination of femtosecond laser at 80 MHz repetition rate is similar to that with CW lasers. This is because of the limited bandwidth of STM feedback electronics, which is typically less than 100 kHz. The tip response due to 80 MHz pulses cannot be detected through normal STM electronics, it is rather than an average effect similar to the results with CW lasers. The transient tip expansion previously measured with AFM cantilever was also limited to the instrument bandwith.[14] It is well known, however, that the time scale of photon absorption is on the order of picoseconds. New experimental technique has to be developed for detecting such ultrafast transient response under the illumination of femtosecond laser pulses.

In summary, the dynamic expansion of an STM tip under illumination of femtosecond laser has been investigated with various parameters. The amplitude of tip expansion is found to be proportional to the incident power. The cut-off frequency has a linear dependence on the illumination spot. Tip expansion under illumination of visible laser is much more significant due to the tip induced plasmon modes. Relatively small tip expansion under illumination of NIR laser and DUV laser suggests a direction for experiments, where the tip expansion has to be minimized.

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