

Estimation of the cooling times for a metallic tip under laser illumination

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The temperature evolution at the apex of a sharply pointed needle submitted to ultrafast pulsed-laser irradiation was determined using a pump-probe method. The laser pulse acts as a pump pulse whereas the probe pulse is a fast high-voltage pulse. Then cooling times are consistent with a heating zone of a few microns with a laser beam polarized along the tip axis and a spot size of 0.8 mm. © 2006 American Institute of Physics. [DOI: 10.1063/1.2181654]

A wide variety of techniques work on the principle of irradiating a sharply pointed needle with a laser beam. The apex of the needle (or tip) can be used for instance as a local probe to scan surface properties. The scanning near-field optical microscope uses the enhanced electromagnetic field produced at the tip apex to obtain a subwavelength resolution.¹ In photoassisted scanning tunneling microscopy (STM), the laser beam is used to investigate optically excited vibrational and electronic states.² The laser assisted field electron microscope uses a negative voltage on the tip to produce emission of electron from the surface of the tip. The use of a laser may produce photoinduced electrons.^{3,4} When applying a positive voltage it is also possible to emit directly the specimen atoms as ions. This is the basic principle of laser-assisted atom probe tomography.^{5,6}

A controversy exists on the physical principles which give rise to the observed interesting phenomena. Indeed, when focusing a laser beam to the apex of a metallic specimen, two consequences are generally accepted: first, a part of the laser energy is absorbed by the specimen generating a temperature increase.^{5,7} This temperature rise occurs whatever the duration of the laser application. Second, if the laser field is polarized along the specimen axis, the sharply pointed shape of the specimen produces a strong enhancement of the electromagnetic field induced by the laser at the apex of the specimen.¹

In STM, the more dramatic consequence of heating is the thermal expansion of tip. This heating is thought to be the main cause of the nanostructure formation at the scanned material surface.² However, some authors suggest that the underlying mechanism is connected with the enhancement of the electromagnetic field at the tip.⁸

Previous work with a laser-assisted atom probe using ultraviolet nanosecond pulsed lasers suggested that the main mechanism of evaporation is the sharp temperature rise following the pulse,^{5,6} but it was indicated that the electric field should be taken into account when using ultra short laser pulses.⁵ Furthermore, it was recently proven that the field evaporation is closely related to the polarization of the laser along the tip axis, which suggests that the induced local electric field is of great importance in the field evaporation phenomenon.⁹

The problem in solving this discrepancy is that both physical effects are difficult to measure in practice since they

appear in a very small region of the specimen (the tip apex). Different models have been developed to evaluate the enhancement of the electric field at the tip extremity.¹ These models indicate that enhancement factors up to several thousands could be reached. However, such enhancements are strongly dependent of both the geometry and the chemical nature of the tip. The problem of the temperature rise is also not so simple. In the simplest approach, the light absorption by the tip can be treated by assuming that only light obstructed geometrically by the tip is absorbed with a certain coefficient. This type of approximation was used by different authors on tips of various geometries.^{5,7} However, diffraction effects must be taken into account, as the dimensions of the tip are of the order of the optical wavelength or below. Indeed, if we consider Fresnel's equations, one naively expects light polarized along the cylinder-axis to be less absorbed. This is true only if the radius of the cylinder is greater than 0.2λ . Taking into account antenna effect, with radius lower than 0.1λ , the parallel component exhibits a greatly enhanced absorption.

In this letter we report the use of an original method to determine the temperature evolution at the extremity of an atom probe tungsten tip. This method is based on the use of a pump-probe scheme.

In atom probe tomography, a tip-like-shape sample is ablated atom by atom by means of an electric field F of a few tens of volts per nanometer. Because the tip radius R is below 100 nm, this huge electric field can be obtained with tip voltages V in the 5–20 kV range ($F=V/\beta R$ with β as a geometric factor). Atoms removed from the surface of the material are projected onto a position sensitive detector where they are chemically identified by time-of-flight mass spectrometry. The time-controlled field evaporation of atoms is ensured by nanosecond high-voltage (HV) pulses superimposed on the standing voltage V_{dc} or by laser pulses of several microjoules. These data are used to obtain a three-dimensional map of the atomic positions in the probed volume with a near atomic resolution.⁵

The instrument used in this study is a linear atom probe with a flight length of 19 cm. The open area of the detector is 46 mm. The experiment is performed under ultrahigh vacuum ($\sim 10^{-9}$ Torr). A 1 kHz pulsed Ti:sapphire laser ($\lambda=780$ nm) with 120 fs pulse duration and tunable energy of up to 1 mJ/pulse was used. The laser beam was slightly focused onto the tip with a spot diameter of 0.8 mm controlled by a charge coupled device camera. The width of time-of-flight peaks is found much better than half a nano-

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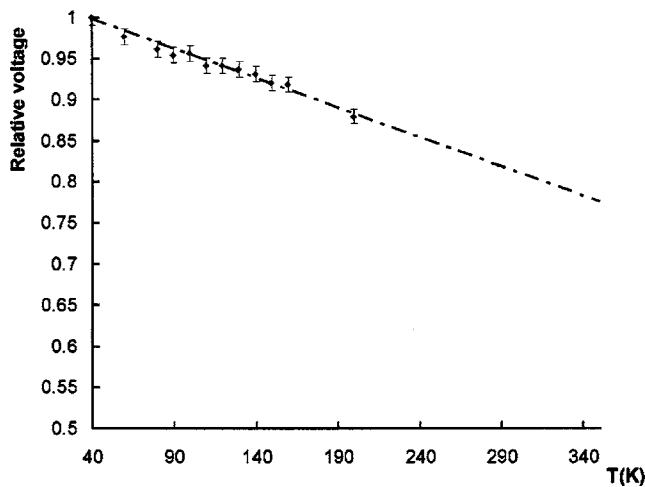


FIG. 1. Relative evaporation voltage as a function of emitter temperature for a tungsten sample. The detection rate is 0.005 atom/pulse.

second, which proves that after 1 ns the phenomenon of evaporation is finished.

Physical theories of field evaporation are based on simple one-dimensional models of atomic and ionic forces, image potential, effects of induced particle polarization. The presence of F at the surface makes ionic states more stable than atomic states as the distance from the surface increases. The change from atomic to ionic state is possible because of thermal vibrations that allow an atom to overcome the potential barrier formed by the crossing of the two potential energy curves. In the HV pulsed atom probe, the pulse amplitude V_p is set in order to generate an electric field F that lowers the potential barrier height during a short time. The probability P to field evaporate an ion follows a Maxwell-Boltzmann law $P = \nu \exp[Q(F)/k_B T]$ with ν as the Boltzmann constant, and T as the tip temperature. This probability is thus very sensitive to F and can be accurately adjusted via the total voltage $V_T = V_{dc} + V_p$ applied to the tip.

Assuming a given probability of evaporation, there is a strong dependence between the electric field at the tip surface and the tip temperature. Figure 1 presents the variation of the voltage required to evaporate a tungsten specimen at a 0.005 atom/pulse detection rate relative to the voltage required at 40 K. To obtain this curve, a tungsten tip with an end radius ~ 30 nm and a very low shank angle was cooled down to 40 K. The total voltage ($V_{dc} + V_p$, with $V_p = 0.2V_{dc}$) was adjusted until the required detection rate was established. The temperature was then incrementally increased and the voltage was adjusted until the same evaporation rate was achieved. The end radius was found constant all over the experiment. Indeed, the evaporation voltage at 40 K was found approximately the same at the end of the experiment than at the beginning. It indicates that the specimen shank angle is very low. Moreover, a limited amount of materials was removed during the whole experiment so that no tip blunting correction is required. As observed experimentally by different authors,^{5,6} the relationship is linear on a wide range of temperature (40–300 K). This curve gives a direct calibration of the temperature T of the specimen during the short time of evaporation (< 1 ns). A thermal pulse of about 300 ± 20 K is needed for 20% evaporation field difference. Note that this value is slightly different from the value obtained by Kellogg (400 ± 20 K).⁶ The source of this differ-

ence is not clear, but this suggests that the 300 K threshold measured in this work provides a minimum estimate for the temperature rise required for field evaporation.

The tip used to obtain the calibration curve was analyzed in laser assisted mode. V_{dc} was kept 20% below the threshold of evaporation. The polarization of the laser beam was set along the tip axis where the field enhancement is the highest. The energy per pulse was increased up to ~ 15 mJ/cm² to obtain a constant evaporation (0.005 atom/pulse) of W³⁺ ions.

Using a pump-probe scheme, it is possible to scan the lattice temperature after the laser pulse. The laser pulse acts as a pump that increases the tip temperature due to the absorption of a part of the laser energy. Indeed, considering the radius of the specimen at the apex (~ 30 nm) and the anomalous skin depth in metals (close to 100 nm),⁵ the specimen electrons may be considered heated uniformly in a section of the tip apex. In the first 10 ps (longer than the electron-phonon coupling time), the energy absorbed by the free electrons is transferred to the ion lattice. The problem is then reduced to a simple thermal transfer problem in the one-dimensional geometry of a tip sample. A voltage pulse is used as a probe to scan the temperature at the tip surface in the few nanoseconds following the pulse. The voltage pulse is triggered by a pulse generator which is delayed from the laser pulse start command. This delay δt is adjusted up to ± 200 ns with a jitter of about 1 ns. The voltage pulse $V_{p,L}$ is incrementally increased to evaporate tungsten at a detection rate of ~ 0.005 atom/pulse. Since both the start time ($t_0 + \delta t$) and the energy of ions in HV mode ($V_{dc} + V_{p,L}$) are different from those in laser mode (t_0 and V_{dc}), two different set of peaks well separated in time are observed. $V_{p,L}$ is then compared to V_p , the pulse voltage necessary without laser at 40 K. The total voltage ratio is used to determine the temperature for the given delay.

Note that the temperature 50 ns before the pulse was found equal to the base temperature indicating that just before the laser pulse, the tip is completely cooled down to 40 K. A strong temperature rise was found after the laser pulse. However, the temperature in the first nanosecond after the pulse was measured well below the temperature required to evaporate in thermally assisted mode. Once the energy was deposited into the tip, the kinetic of temperature simply follows the diffusion equation of the heat conduction $\partial T / \partial t - \alpha(T) \nabla^2 T = 0$, with $\alpha(T)$ as the thermal diffusivity of the conduction medium. An analytical solution to this problem is found if we assume the tip as a simple semi infinite cylinder along the x tip axis, with a heated region described by a Gaussian formula ($T = T_0 + T_{max} e^{-(x^2/2\sigma_L^2)}$, with σ_L as the size of the heated region). Since the measured temperatures are between 100 and 200 K, the thermal diffusivity was taken constant in a first approximation ($\sim 9 \cdot 10^{-5}$ m² s⁻¹, the value expected at ~ 150 K).⁷ Note that radiation effects were assumed negligible at these temperatures. The temperature relaxation is given by $T(x, t) = T_0 + T_{max} / \sqrt{[1 + 2(\alpha t / \sigma_L^2)]} \cdot e^{[x^2/2(\sigma_L^2 + 2\alpha t)]}$. This equation fits remarkably well the cooling observed experimentally with $T_{max} = 160 \pm 20$ K and $\sigma_L = 1.5$ μ m (Fig. 2), strongly smaller than the laser spot size onto the tip (~ 800 μ m). This discrepancy is not due to approximations done to compute the temperature relaxation. Indeed, more refined models which take into account the actual variation of the diffusivity with

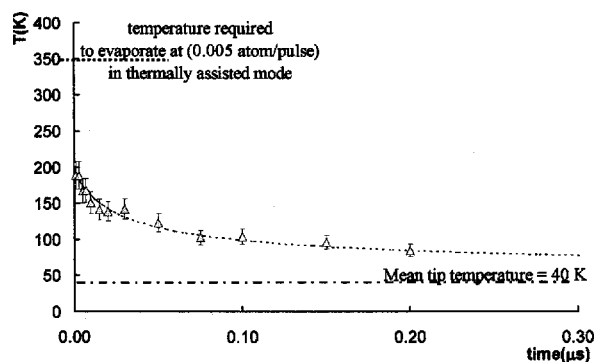


FIG. 2. Variation of the temperature after the laser pulse for a laser polarized along the tip axis (triangles, 15 mJ/cm^2). The temperature relaxation is fitted by the model defined by $T(0,t) = T_0 + T_{\text{max}} / \sqrt{1 + 2(at/\sigma_L^2)}$ with $T_0 = 40 \text{ K}$, $T_{\text{max}} = 160 \text{ K}$, and $\sigma_L = 1.5 \text{ } \mu\text{m}$.

temperature and the complex geometry of the tip showed the same temperature variations for heated regions of a few microns. This result strongly suggests that the component of electromagnetic field aligned with the tip axis contributes to heat only the last few microns of the very apex of the tip, where diffractions and field enhancement effects are the highest. Indeed, only this last part of the tip has a diameter that is smaller than the laser wavelength. The original method presented in this letter, based on a pump-probe scheme, enables the determination of the local lattice temperature at the nanosecond scale after the laser pulse. If the extrapolation to

$t=0 \text{ ns}$ is correct it would indicate a peak temperature rise of only 160 K, far less than the minimum temperature of 300 K expected for field evaporation, suggesting that the process is not purely thermally activated, but that there is a direct field component. However, the only uncertainty concerns the lattice temperature evolution in the first picoseconds after the pulse. Indeed on the nanosecond time scale, it is clear that the temperature across the tip width is homogeneous, so that the one-dimensional model is valid. However, on the picosecond time scale a nonhomogeneous temperature may be found between the surface and the core of the specimen especially far from the apex where the diameter is larger than the mean free path of electrons. The time constant for decay in the picosecond scale can therefore be different. The hypothesis of strong temperature variation in the first nanosecond cannot be dismissed.

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