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Effect of the laser pulse width on the field evaporation behavior of metals and oxides

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ABSTRACT

The laser assisted field-evaporation of metals and oxides strongly depends on the illumination conditions. Here we study the effect of laser pulse width using two different laser systems, with a pulse duration of a few tens of femtoseconds and a few tens of picoseconds, respectively. Adjusting the laser wavelength by nonlinear optical conversion systems, we study the evaporation behavior of FeCu and MgO samples. We prove that the laser pulse width does not affect the evaporation behavior, in the range of duration explored. These results are explained taking into account the absorption behavior of nanometric samples and their thermal properties.

1. Introduction

In Laser-assisted Atom Probe Tomography (La-APT), laser pulses are used to trigger the field evaporation of surface atoms [1]. This technique, able to image materials with 3D subnanometric resolution, is currently being more and more applied in different domains of nanoscale science and technology [2–6].

To optimize the performance of the La-APT analysis, the illumination conditions have to be adjusted to the analyzed material. Indeed, in La-APT, surface atoms are removed one by one by field emission from sharp tips with an end radius of around 50 nm subjected to a high dc field of several tens of volts per nanometer; the interaction of the laser beam with the tip increases its temperature and hence allows the evaporation of surface atoms [7–9]. This temperature increase is related to the light absorption at the nanometer scale. It was proved that the laser wavelength, polarization, repetition rate and the tip shape can influence the laser–tip interaction and, hence, the ejection process [10–14]. However, up to now, no systematic study on the role of laser pulse duration has been reported [15]. Usually, laser pulses of 50 fs, 500 fs or 30 ps are used. It is well known that the laser–matter interaction strongly depends on the pulse duration, as widely studied in the domain of surface laser ablation. Non-thermal processes, such as coulomb explosion, are reported for short pulses and thermal behavior is shown to depend on laser pulse duration [16–19].

In this paper, we study the influence of the laser pulse duration using two difference laser systems to trigger the evaporation: an ultra-fast fs laser (50 fs) and a ps fiber laser (30 ps). We prove that the evaporation process can be triggered by picosecond light pulses from a monolithic fiber laser system. Ultra-fast fiber lasers have attracted much interest in recent years due to their inherent advantages such as compactness, stability and turnkey operation. Driven by several industrial and scientific applications such as material processing, nonlinear microscopy, and precision metrology, the output power of ultra-fast pulsed fiber laser systems has undergone an impressive growth during the past decade [20]. In particular, the exploitation of large mode area photonic crystal fibers in chirped-pulse amplification schemes has permitted the achievement of record pulse energies of several millijoules at high repetition rates [20]. However, the non-compatibility of these rod-type fibers with step-index fiber components and the need for gratings-based stretchers and compressors make these systems highly bulky and relatively complex. So, the development of high power ultrafast laser systems in all-fiber format with performances qualified for scientific and industrial applications is still challenging. Here, we report the development of an alignment-free high-power fiber laser and demonstrate its use for atom probe analysis. This first experimental proof of ion evaporation assisted by fiber laser is a first step towards the development of an alignment-free all-fiber optical system (from the laser to the focusing system in the high vacuum chamber), which will ensure a high stability of the working conditions. We analyze two different kinds of samples: metallic sample with an extremely low thermal diffusivity and wide-band-gap oxide sample.

In the case of metallic samples, it was proved that the evaporation is thermally assisted by the absorption of the laser energy

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[7–9]. Hence, we study the effect of the pulse duration on the heating process of the sample. We choose low-diffusivity materials, because for this kind of metallic tips, the illumination conditions were shown to be critical and have to be adjusted according to the sample shape [13,14].

In the case of wide-band-gap oxides, non-thermal features were indeed identified. However, non-thermal excitations of a wide-band-gap tip take place only at the surface and are believed to influence evaporation only during first several picoseconds [21]. The role of non-equilibrium processes in laser-assisted field evaporation of semiconductors is under investigation and will be published elsewhere. Concerning the longer evaporation process, it is proved to come from thermal effects in the whole volume of the tip [22]. In fact, it was recently reported that the absorption can be separated in two contributions: a surface absorption, strongly influenced by the dc field, and a volume absorption related to the optical properties and geometry of the sample [22]. The surface absorption is enhanced by the band-gap shrinkage under high dc field and it is mainly a linear optical process [23,24]. The volume absorption, in wide-band-gap materials, is proved to be a multi-photon process, hence strongly dependent on the laser pulse duration [22,25]. The results reported in this paper show that, contrary to what expected, the two processes are scaled in the same way with the pulse duration.

The paper is organized as follows: in the first section, we present the experimental setup, the sample preparation methods and the laser systems used. In the second section, our experimental results are presented and discussed, studying the laser–nanotip interaction numerically. Eventually, in the last section, our conclusions are drawn.

2. Experimental section

In our experiments, the specimen is placed into the ultra high vacuum ($<10^{-7}$ Pa) chamber of a tomographic atom probe with a flight path of about 20 cm. A position-sensitive detector (PSD) [26] with improved multi-hit capabilities is used to accurately measure the mass spectrum of evaporated ions, by time-of-flight measurement. A fast photodiode is used to accurately measure the moment at which a laser pulse is sent onto the tip. This signal is used to trigger the time-of-flight measurement channel. In order to ensure a good stability of the laser power, the whole electronic device is synchronized with the internal clock of the laser.

2.1. Sample preparation

Metallic tips of FeCu are prepared by eletro-polishing method using micropolishing method [27] with a platinum wire loop (diameter of 2–3 mm) containing a small quantity of an electrolyte (a mixture of 98% of monobutilic ether and 2% of perchloric acid). Oxide tips of MgO are prepared by focused ion beam (FIB) milling [28]. The annular pattern FIB milling with 30 keV Ga beam acceleration is used until achieving a tip radius of curvature of 75 nm. The damage produced by the 30 keV beam is removed using a 2-keV beam, reducing drastically amorphization and implantation effects.

2.2. Laser systems

The femtosecond laser system used is a 1-kHz pulsed Ti:Sa laser ($\lambda = 788$ nm) with 35-fs pulse duration and a tunable energy of up to 2.5 mJ/pulse (Spectra-Spitfire Pro XP). An optical parametric amplifier from Light Conversion, TOPAS-C has been recently implemented at the output of the laser to tune the wavelength from 200 nm to 800 nm.

The picosecond laser consists of a home-made passively mode-locked ytterbium-doped fiber laser operating in the all-normal dispersion regime [29]. A fiber-Bragg grating-based filter centered at 1040 nm is included inside the cavity to limit the spectral bandwidth to less than 140 pm [30]. The laser generates nearly transform-limited pulses with 30 ps duration and less than 1 nJ energy, at a repetition rate of 8 MHz. A two-stage fiber master-oscillator power-amplifier (FMOPA) is used to boost pulse energy to more than 300 nJ. This corresponds to more than 2.5 W of average power. Laser emission is then frequency doubled in a nonlinear crystal (BBO) to produce few tens of nanojoules energy at 520 nm wavelength. An electro-optics modulator is used to lower the repetition rate to 1 kHz which is a more adapted triggering frequency for mass spectroscopy measurements. The laser beam is focused onto the tip with a spot diameter of a few μm controlled by a CCD camera. The laser beam linear polarization is adjusted parallel to the tip axis using a half-wave plate.

3. Results and discussion

3.1. FeCu sample

The same FeCu tip was analyzed in APT using the fs laser at the fundamental wavelength of 788 nm and then the fiber laser at the fundamental wavelength of 1040 nm. The voltage applied to the sample was fixed at 7 kV, the laser power was increased in order to obtain the evaporation rate of 0.02 ions/pulse and a laser contribution of 10%. The obtained mass spectra are reported in Fig. 1 (a).

The two spectra are well superimposed, and no difference between the peak widths at half of the maximum (FWHM) or even at ten of the maximum (FWTM) can be measured. The FWHM of the mass peaks is related to the ion evaporation time. For a thermally assisted evaporation, the evaporation time is related to the cooling time of the tip by $\tau_{evap} = 0.1\tau_{cooling}$ [31]. The cooling time $\tau_{cooling} = w^2/D$ is the characteristic cooling time of a nanowire

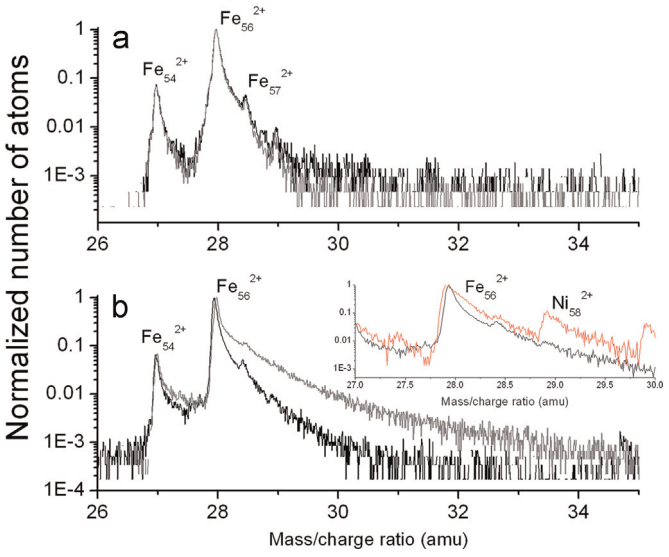

Fig. 1. Normalized mass spectra of FeCu tip: (a) gray line using 50 fs laser at 788 nm; black line using 30 ps laser at 1040 nm and (b) gray line using 50 fs laser at 394 nm (SHG); black line using 30 ps laser at 520 nm (SHG). Inset: Normalized mass spectra of a Fe-based alloy tip using 120 fs laser at 500 nm from Ref. [10](red line); Normalized mass spectra of FeCu tip using 30 ps laser at 520 nm (SHG) (black line). (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)