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Nanomilling surfaces using near-threshold femtosecond laser pulses

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Abstract. We have produced crater depths of less than 10 nanometers using 100-1000 pulses of a near-infrared femtosecond laser (800 nm, 125 fs) on a copper thin film surface. By determining the single-shot ablation threshold, incubation coefficient and surface reflectivity, the femtosecond laser pulse parameters for surface nanomilling are established close to the multiple-pulse ablation threshold limit for a copper thin film. Photomultiplier measurements of a copper emission line were used as a real time monitor of the nanomilling process for which photons were detected only once every several shots. The results are consistent with a model that ablation occurs in bursts every several shots after a number of intervening incubation energy storage shots.

1. Introduction

High precision surface nanomilling of metals using femtosecond laser pulses requires investigation into the material response resulting from multiple pulses at near-threshold energy densities. We show that surface material removal at intensities near the ablation threshold can be controlled to remove material at an effective rate of picometers per laser shot using near-infrared femtosecond laser pulses. Controlling the removal parameters at the nanometer level allows for the application of lasers in a new field of surface nanomilling with potential applications in the fine tuning of feature thicknesses and response of microelectromechanical (MEMS) devices, surface acoustic wave (SAW) devices and photonic structures in integrated optical circuits. We are currently investigating the control parameters that lead to removal of single nanometer scale layers with little subsurface damage.

2. Experimental Procedure

The experimental setup, laser equipment and techniques were similar to that used to establish the single and multiple shot damage thresholds in copper foils [1]. In this work, a 15 cm BK7 biconvex lens was used to focus the laser onto the sample surface. A photomultiplier tube (PMT) (Hamamatsu R7518) collected light over a solid angle of $\sim 6 \times 10^{-3}$ Sr and a 326 nm (10 nm bandwidth) interference filter was placed in front of the tube to measure emission from the 325 nm and 327 nm lines of excited copper atoms that are ejected from the sample during ablation. The PMT was calibrated using a diffuse scattering barium sulphate plate irradiated with calibrated laser pulses to provide a known intensity source (W/Sr) with an absolute error of $\pm 15\%$. Each PMT trace generated by a laser shot was saved and numerically integrated over a 25 ns window to obtain integrated current, which is proportional to the number of photons observed.

The average laser pulse width was 125 fs full width at half maximum (FWHM) assuming a Gaussian pulse shape and varied over a range of 121 fs to 127 fs during the experiments. The pre-pulse contrast ratio was 3×10^{-4} to 9×10^{-4} measured after a half-wave plate and Glan polarizer combination, which was used for control of the experimental laser energy. The contrast ratio varied within this range according to the setting of the half-wave plate due to different polarizations of the pre-pulse and main pulse. Further energy selection was made using Schott NG absorbing glass filters. The repetition rate of the laser, maximum 1 kHz, was set between 1 Hz and 50 Hz to minimize counting errors. Nanomilling experiments with the PMT were performed at 1 Hz.

Different copper thin film samples sputtered on silicon wafers were used throughout this study with thicknesses of 100 nm and 300 nm. The single-shot ablation threshold, $\phi_{th}(1)$, incubation parameter, ξ , and reflectivity, R , were measured for each sample as described in [1]. The $\phi_{th}(1)$ for the 100 nm and 300 nm copper thin film samples were measured to be (852 ± 37) mJ/cm² with a reflectivity of (97 ± 1) % and (896 ± 80) mJ/cm² with (83 ± 1) % reflectivity, respectively, at the 800 nm wavelength. Both thresholds deviated from the single-shot ablation threshold previously reported for a bulk copper sample and a thin copper film with a 250 nm thickness where $R \sim 95$ % [1]. It has been reported that $\phi_{th}(1)$ for thin films of metals will decrease as the film thickness approaches the characteristic length for electron diffusion [2-4]. The lower $\phi_{th}(1)$ for the 100 nm copper film is within this expectation. The $\phi_{th}(1)$ for the 300 nm thin copper film was anomalous, but given the low reflectivity it probably has an altered crystal structure from pure polycrystalline copper as a result of the deposition conditions employed for fabrication.

Despite the discrepancy in the $\phi_{th}(1)$ for the 300 nm thin copper film, the incubation coefficient of this sample agreed with the value previously reported [1]. The average incubation coefficient for both samples was measured to be 0.77 ± 0.01 . We believe this makes the 300 nm sample an interesting comparison to our well-controlled thin copper film samples. The incident fluence for each experiment was established using the Gaussian beam limiting technique by measuring the diameters created by the single shot ablation craters versus energy and extracting the beam waist for each experiment [1, 5].

3. Results and Discussion

Optical profilometry scans (Zygo New View 5000) of the nanomilled surfaces established that nanomilling occurred near the ablation threshold for copper. Figure 1 shows nanostructured profiles for copper thin film surfaces as they occurred with respect to the ablation threshold and incident pulse number. All profiles were plotted with the same aspect ratio of 30 μ m:10 nm (3000:1) making all data directly comparable except for that labelled *A*, which is the 83 % reflective sample. Using

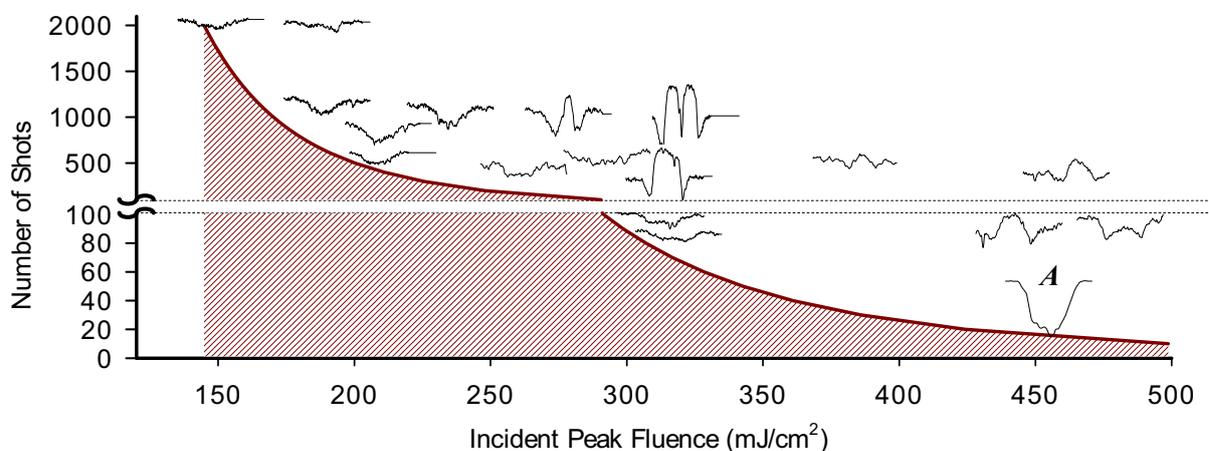


Figure 1. Nanostructured surface profiles measured by optical interferometric profilometry. The scan length in each profile is 30 μ m across and all aspect ratios are equivalent at 3000:1 and directly comparable, except for the profile labeled *A*. This profile is discussed in the text.

$\phi_{th}(1) = 852 \text{ mJ/cm}^2$ and $\xi = 0.77$, a line is drawn in figure 1 to differentiate between the regions expected to show material removal and the region where no damage is expected.

All crater profiles given in figure 1 were less than 10 nm with the exception of the profile labelled *A* (83 % reflective sample). Structures produced very close to the nanomilling threshold were not perfectly Gaussian in shape and may indicate a nonlinear enhancement of minor irregularities in the beam. Incident energy densities farther above the incubation line resulted in surface structures with bumps in the centre of the laser interaction region rising to heights of less than 10 nm. Although surface nanostructuring may be of interest, our goal was to avoid such structures in our final results. These nanostructures may be void formation within the sample or between the film and the substrate. To differentiate between a purely mechanical failure and thermal failure, nanomilling experiments with polished bulk copper samples are planned to help fully quantify the nanomilling process and avoid mechanical failure of the thin film. These issues are important as the proposed application is towards thin copper film devices.

Reflectivity played a significant role in the nanomilling results in the multishot regime as well as the single shot. The nanomilled shot indicated by *A* in figure 1 showed a hole 371 nm deep milled through to the silicon substrate by illuminating the 300 nm copper thin film sample with a reflectivity of 83 % with 20 pulses at a peak incident fluence, ϕ_0 , of 458 mJ/cm^2 . The enhanced drilling rate is not surprising if the incident energy is scaled with reflectivity except that *a priori* knowledge of reflectivity may not be available when operating on specific devices under test, making online monitoring a key component in nanomilling.

To establish a simple real time monitor of the nanomilling process, a PMT was used to detect optical emission from ejected species during the ablation process. An interference filter was used to filter out the background radiation signals from the laser light and blackbody emission. Figure 2(a) shows an example of the PMT trace measured for the sample irradiation leading to profile *A* in figure 1. In contrast, irradiation of the sample with 97 % reflectivity illuminated with 500 pulses having a ϕ_0 of 290 mJ/cm^2 yielded a crater of 1.9 nm depth and PMT signals as shown in figure 2(b). For the 371 nm hole, the PMT measured many photons emitted at every shot while for the 1.9 nm hole, two spikes of approximately the same integrated values were measured. These spikes in figure 2(b) correspond to single photon detection events. We estimated that the probability of observing a single false positive event in 1000 laser pulses was less than 1 % making the observation of two photons emitted in a run of 500 shots statistically significant. The result shows that irradiation at the low fluence of 290 mJ/cm^2 leads to very gentle ablation with little excitation of the copper atoms. The average material removal rate corresponds to a few isolated atoms being removed by each

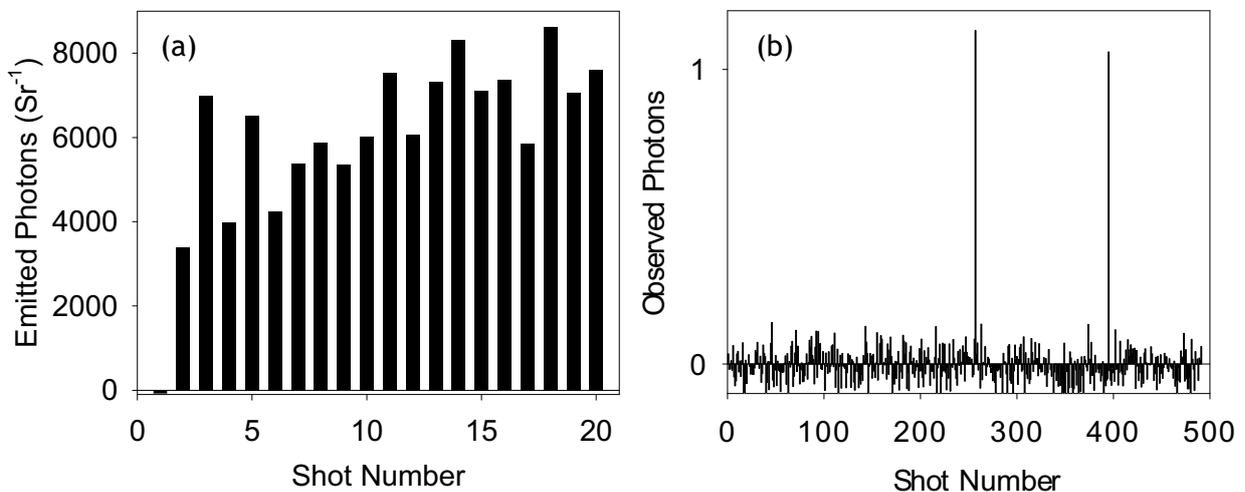


Figure 2. PMT traces of nanomilled results for the (a) 83 % reflective sample at $\phi_0 = 458 \text{ mJ/cm}^2$ and (b) 97 % reflective sample at $\phi_0 = 290 \text{ mJ/cm}^2$.

laser pulse, a process observed in molecular dynamics (MD) simulations [6]. Nanomilling could also occur in bursts with energy being stored in the sample over many sub-threshold shots and released in an ablation event after a certain level of stored energy has been reached. Such a mechanism would be consistent with the isolated photon emission events shown in figure 2(b).

A key component to nanomilling is the incubation of damage due to the absorption of laser energy in the sample. MD simulations with silicon in our lab show that irradiation with sub-threshold 100 fs pulse energies leads to modification of the material lattice 30 ps after the termination of the laser pulse. Any modifications from the equilibrium lattice structure means that there has been energy stored in the sample that can be liberated in subsequent irradiation events.

It is expected that the excitation of copper atoms was a stochastic event that occurs with a certain probability increasing with the absorbed fluence. The number of emissions observed could be used as a diagnostic for the rate of the nanomilling process. An online monitoring system such as the PMT would indicate when the process was progressing as expected.

4. Conclusion

It has been demonstrated that near-threshold femtosecond laser pulses in the near-infrared wavelength region (800 nm, 125 fs) can nanomill copper surfaces to depths of less than 10 nanometers. Effective ablation rates were picometers per pulse corresponding either to a few scattered atoms being removed in any given laser shot or to storage of energy over a number of irradiation cycles with a release in a single ablation event. It has also been shown that a PMT monitor can measure the ablation rate due to the occasional excitation of an ablated copper atom. Further detailed scaling studies will be required to determine whether the nanoablation is occurring continuously or occurs as discrete events.

5. Acknowledgements

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