Nanoproccessing in the Near-Field
of Atomic Force Microscope Tip by Femtosecond Laser Pulses

A. Kirsanov, A. Kiselev, <u>A. Stepanov</u>

**Institute of Applied Physics of Russian Academy of Science, 603950, 46 Ulyanov st.
Nizhny Novgorod, Russia, 831 216 4907, step@ufp.appl.sci-nnov.ru**

N. Polushkin

**Institute for Physics of Microstructures of Russian Academy of Science, 603600, Nizhny
Novgorod, GSP-105, Russia, 831 267 5313**

The formation of nanostructures on the surface of different materials is a field of activity that attracts much attention in recent years. One of the methods to produce nanostructures is to illuminate a gap between a tip of atomic force microscope (AFM) or scanning tunneling microscope and the substrate surface by a laser radiation. In such a way hillocks and pits with a diameter down to 20-30 nm on the surface from different materials have been created [1-3]. For explanation of nanostructuring observed two mechanisms are discussed in literature. One mechanism is the substrate material ablation by a laser electric field enhanced in the vicinity of the tip apex [1, 4]. Another one is a mechanical stress by a heated tip as a result of thermal expansion of a tip due to absorption of laser radiation [3, 5, 6]. Despite of large efforts made to clarify the problem a controversy in a nanoproccessing explanation still exists. In this letter we present the results of several experiments performed with laser pulses of femtosecond duration in order to contribute further to the discussions in mechanisms of nanostructuring. The use of ultrashort laser pulses introduces a new parameter (pulse duration) important for better understanding of the problem. In addition it occurred that femtosecond duration of the laser pulses allows to enlarge a circle of materials, that can be developed.

The radiation from a femtosecond laser (λ=0.8 mkm, τ=100 fs, $W \le 50$ mkJ) is focused by a lens to a focal spot of 300 mkm in diameter in a tip – surface gap. Angle of incidence is $\theta = 17^0$ relative to the substrate surface, we change the polarization of laser radiation during the experiments. A commercial AFM system (Smena model from NT-MDT) is used. Silicon tips covered by 20-30 nm layer of W_2C with a radius of curvature $r \approx 30$ nm are used. The samples are made of metallic films (In, Au, Cu, magnetic material FeCr) with a thickness of 20-30 nm on a Si substrate. The films are deposited by sputtering technique. The scanning probe microscope is mainly used in a contact mode.

When the fluence is above a certain threshold value, processing of the craters on the surface of the film occurs. Fig. 1 shows a typical crater on FeCr film produced by a pulse with a fluence F=95 mj/cm^2 taken by an AFM immediately after processing. The craters are typically 30-50 nm in a diameter with 3-10 nm in height similar to the results of Jersch [1] and Lu [7]. The threshold fluence depends on the material of the film. In Fig. 2 the dependence of the threshold fluence on the melting temperature for different materials of the sample films is presented: it is approximately linear. It is observed that the threshold fluence F_{th} is influenced by a polarization of laser radiation. When p-polarization of the laser with respect to the substrate surface is used the value of the threshold fluence is 1.5–2 times lower, then for perpendicular s-polarization. Thus for FeCr F_{th}^{p}=75$\pm$10 mJ/cm^2, F_{th}^{s}=150$\pm$15 mJ/cm^2, for Au F_{th}^{p}=34$\pm$5 mJ/cm^2, F_{th}^{s}=67$\pm$10 mJ/cm^2.

The comparison of the threshold fluence for femtosecond and nanosecond laser pulses is performed. To produce nanosecond pulses regenerative amplifier of the laser system is used in a regime of lasing (without femtosecond pulse injection). The threshold fluences F_{th}^{p}=46$\pm$5 mJ/cm^2 and F_{th}^{s}=115$\pm$10 mJ/cm^2 for nanofabrication on a gold film are measured with a pulse
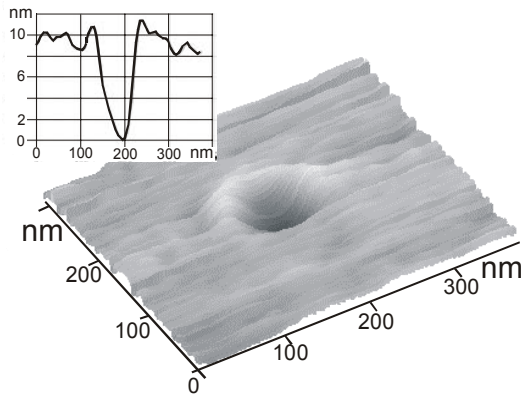
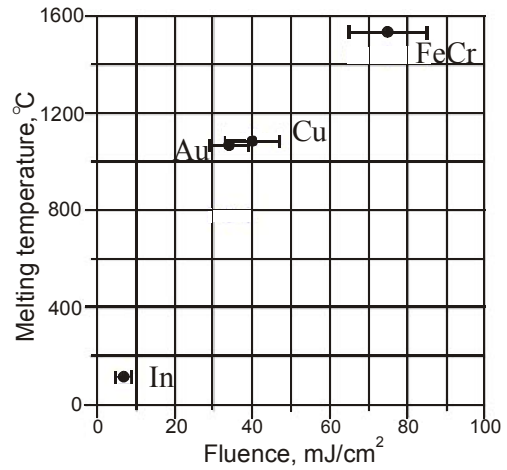Fig. 1. Image of a crater on FeCr, *p*-polarization, F=95 mj/cm^2

Fig. 2. Threshold fluence vs melting temperature

duration τ=9 ns (as compared with F_{th}^{p}=34$\pm$5 mJ/cm^2 and F_{th}^{s}=67$\pm$10 mJ/cm^2 with femtosecond pulses).

The dependence of the threshold fluence for FeCr film on the distance between tip and sample surface for *p*-polarization of laser radiation is depicted in Fig. 3. This dependence is practically absent within the experimental error. The relationship between the depth of nanostructures and the pulse number is shown in Fig. 4.

As was mention in the introduction two main mechanisms for nanoproccessing: the ablation of sample material by a laser electromagnetic field enhanced at the tip apex and the mechanical stress from the tip heated by a laser are under discussion till now. Let us consider our experimental results from the point of view of these mechanisms. Field enhancement in the vicinity of the tip apex is very sensitive to the polarization of the laser [8]: if the enhancement in the intensity up to several hundred times is calculated for *p*-polarization, only minute variation in intensity for *s*-polarization is predicted. The tip heating is determined by laser radiation absorption [3, 6]. The absorption averaged over the conical surface of a W tip (Fresnel formulas are taken into account) is presented in Fig. 5, $\psi = 0^0$ corresponds to *p*-polarization, $\psi = 90^0 - s$-polarization: the difference in absorption does not exceed 10%. Our experiments demonstrate a modest influence of a polarization on the threshold fluence, much less pronounced, then field enhancement mechanism implies.
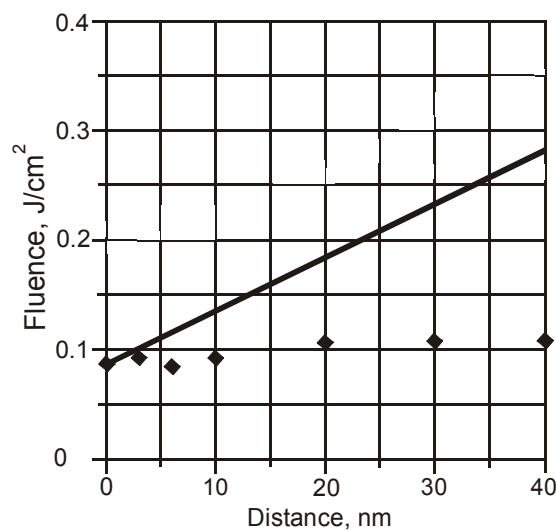

Fig. 3 Threshold fluence vs tip-sample distance for FeCr, *p*-polarization, solid line – mechanical stress model.
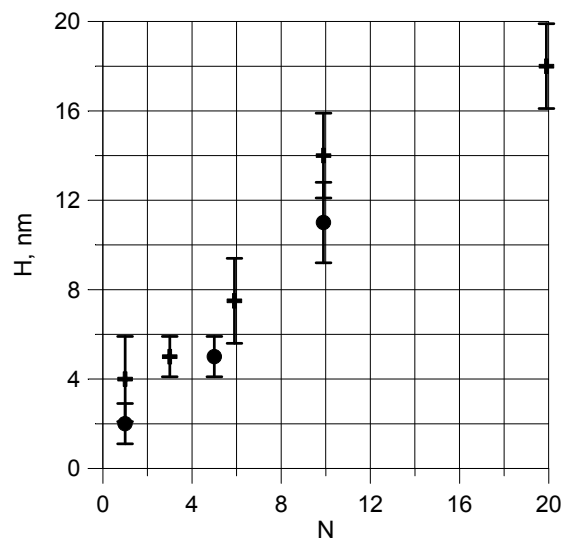

Fig. 4. Depth of nanostructures as a function of number of pulses for Au, crosses – *p*-polarization, F=38 mj/cm^2, circles – *s*-polarization, F=55 mj/cm^2
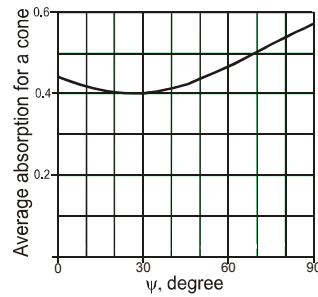
Fig. 5. Average absorption for a W cone as a function of laser polarization

The ablation of the sample material from under the tip should be strongly affected by pulse duration. The absorption of the laser radiation occurs by the electrons in the region of enhanced field with a diameter comparable with a tip apex diameter [8]. Due to heat transport the absorbed energy spreads in a thin metallic layer over the space scale $l \sim \chi\,\tau$ where χ is a coefficient of thermo conductivity and τ is a characteristic time. For the femtosecond duration of the pulses characteristic time is determined by the time necessary to transfer the absorbed energy from the electrons to the lattice τ_l (a few ps). In a case of ns pulses - $\tau \approx \tau_0$ - duration of the pulse, or in other words ns pulse heats a volume approximately $(\tau_0/\tau_l)^2 \sim 10^6$-$10^8$ larger than fs pulse, which should lead to much higher threshold fluence for ns pulses. This is in a strong contradiction with our experimental results. The heating of the tip depends only on absorbed energy, but not on pulse duration (until the heat spreading along the tip occurs). Only small reduction of the temperature of the very tip apex is expected for time interval in the order of 10 ns [6]. This is an explanation of the experimental fact why the threshold fluence is only 1.5-2 times higher for ns pulses than with fs pulses.

The dependence of the threshold fluence on the tip – sample distance should be different for two mechanisms that are under discussion. The enhanced field drops over exponentially with a distance from the tip apex [8, 9], thus the threshold fluence for nanoproccessing should rise sharply with a distance increase. On another hand the heating of the tip does not change when the tip – sample gap changes in several tens of nm. The mechanical stress the tip introduces to the sample is proportional to the difference between the tip lengthening due to heating and tip – sample distance which leads to the linear dependence of the threshold fluence on the tip-sample gap in this model (solid line in Fig. 3). If nanoproccessing takes place when the melting of the film material occurs (this situation is quite probable though additional investigations are required), then no dependence of the threshold fluence on the tip – sample distance should be. In any case the dependence should be weak for a mechanical stress by a heated tip mechanism in agreement with experimental results presented in Fig. 3.

In conclusion, the formation of nanocraters on the metallic films under the atomic force microscope tip by the irradiation of femtosecond laser pulses is demonstrated. Craters with diameters down to 20-30 nm on the surface of wide circle of metals including hard materials with high melting temperature are produced. Experiments devoted to the clarification of the reason standing behind the material development are performed. The analysis of the experimental results implies that the mechanical stress of the AFM tip heated by a laser is responsible for the nanoproccessing on the metallic films investigated in our experiments.

1. J.Jersh, K.Dickmann, Appl. Phys. Lett. 68, 868 (1996).
2. S.M.Huang, M.H.Hong, Y.F.Lu, B.S.Lukyanchuk, W.D.Song, T.C.Chong, J. Appl. Phys. 91, 3268 (2002).
3. R.Huber, M.Koch, J.Feldmann, Appl. Phys. Lett. 73, 3521 (1998).
4. A.A.Gorbunov, W.Pompe, Phys. Status Solidi a 145, 333 (1994).
5. V.A.Ukraintsev, J.T.Yates, Jr., J. Appl. Phys. 80, 2561 (1996).
6. V.Gerstner, A.Thon, W.Pfeiffer, J. Appl. Phys. 87, 2574 (2000).
7. Y.F.Lu, B.Hu, Z.H.Mai, W.J.Wang, W.K.Chim, T.C.Chong, Jpn. J. Appl. Phys. 40, 4395 (2001).
8. O.J.F.Martin, C.Girard, Appl. Phys. Lett. 70, 705 (1997).
9. L.D.Landau, E.M.Lifshitz, Electrodynamic of Solids (Moscow, Nauka, 1982), p.46 (in Russian).