Influence of Irradiation Dose on Laser-Induced Surface Nanostructures on Silicon

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Abstract

We report on the dependence of femtosecond laser-induced periodic surface structures on an increase of incident pulse number. On silicon, the patterns evolve from linear, parallel sub-wavelength ripples, grossly perpendicular to the laser polarization, via coalesced wider features parallel to the polarization, to a crater with periodically structured, pillar-like walls. Closer inspection of the patterns indicates, that the different features always continue to exhibit reminiscence to the preceding lower-dose patterns, suggesting that, indeed, all patterns can be created by ONE single GENERAL formation process, as in self-organized structure formation, and the different structures / feature sizes are NOT due to DIFFERENT mechanisms.

Keywords: Laser induced periodic structures (ripples); self-organized structure formation; silicon; femtosecond ablation

1. Introduction

After a first period of intense research in the 1980's [1-3] the formation of laser-induced periodic surface structures (LIPSS) has seen a lively revival in the last decade when irradiation with ultrashort, i.e. subpicosecond pulses brought about a great variety of new features [4-16], such as "nano" ripples (with a feature size significantly smaller than the laser wavelength), "classical" or "micro" ripples (at the order of the wavelength), "macro-ripples" (in a several-microns range), but also even more complex patterns like arrays of conical features. Most often, several of these structures are even observed coexisting in the same laser spot.

Despite the ample experimental work, however, the underlying physical mechanisms are not yet fully understood. So far, there are mainly two, fundamentally different, approaches:

Model (A) involves a lithographic like process, where a modulated input energy distribution, produced by some interference, is transformed into a corresponding ablation pattern and thus results in a modulated surface morphology [2] (Fig. 1 a)). In this case, the irradiated material only plays a *passive* role, since all structure formation is due to a specific irradiation pattern. Model (B) assumes an *active* contribution of the irradiated material. Here, the laser serves as an energy input, perturbing and softening the target lattice. Then, surface structure formation should occur via self-organization during relaxation from this instability [17,18] (Fig. 1 b).

In principle, both models bear their own advantages and drawbacks. The lithographic, or "classical" model (A) is appealing because, very often, the long, almost parallel surface structures resemble very much a typical interference pattern, as well as does the pronounced dependence on the laser polarization. Further, in many cases, their feature size ("wavelength") is close to the laser wavelength. Such structures are often named

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"LSFL" ("low spatial frequency LIPSS") [19,20], to be distinguished from femtosecond-laser induced nano-ripples, also termed "HSFL" ("high spatial frequency LIPSS"). Such nano-ripples constitute, however, one of the major drawbacks of such model: the sub-wavelength feature size rules out any simple interference of the laser with coherently scattered light. To overcome that problem, additional interactions have been introduced, such as an adaptable (by an adjustable density of surface free electrons) refractive index of the surface selvedge layer, the co-action of higher harmonics and, finally, the involvement of high-frequency surface plasmons [16,21,22]. As a consequence, the model strongly distinguishes between nano- and micro- ("classical") ripples and requires DIFFERENT interaction mechanisms. It is, so far, not at all capable to account for even larger structures (macro ripples) or even more complex patterns, such as cones, "bubbles" etc.. Finally, structure formation by circularly polarized irradiation [23] can not at all be explained by this model.

The self-organization model, (B), can account, instead, for most of the observed patterns, qualitatively, at the same time. In contrast to model (A), model (B) can explain the different types of patterns by one SINGLE physical mechanism (cf. Fig. 2). The model also includes the well known pattern change upon increasing the irradiation dose (cf. Fig. 3); either by increasing the laser fluence or by increasing the number of incident pulses (cf. [24] and refs. therein]. Recently, even the strong polarization-dependence could be included in the model, though the individual micro-physical mechanisms are not yet revealed [18]. The main drawback of this model (B) is, however, that despite its strength in QUALITATIVELY reproducing, in principal, all observations, even the very high aspect ratio often observed, it does not yield any QUANTITATIVE predictions, which, in contrast, seems so easy with model (A).

It is the aim of the present paper to provide additional support for model (B), at the same time putting some more doubts on model (A). For this purpose, we study in detail the pattern evolution when the irradiation dose is increased.

2. Experimental

In our experiments, we irradiated conventionally cleaned silicon (100) wafers in ultra-high vacuum (10^{-9} mbar) by ultra-short, linearly polarized laser pulses from an amplified Ti:Sapphire laser ($\tau_{pulse} \approx 120$ fs; $\lambda =$ 800 nm; repetition rate: 1 kHz; focus diameter ≈ 100 µm). At a constant peak intensity of 2.15×10^{12} W/cm^2, slightly above the ablation threshold for silicon ($I_{th} = 2\times10^{12}$ W/cm^2) [25,26], we increased the irradiation dose by increasing the number of pulses per (fresh) irradiated spot.

After irradiation, the ablated/modified areas were investigated in situ by scanning electron microscopy (SEM).

3. Experimental results

A series of typical results is presented in Fig. 4, showing the effect of three different doses, one of 10 pulses, one of 50 pulses, and one of 1,000 pulses. Already at first sight, it is obvious that there is a clear evolution of the patterns. At low dose (panel a)) the whole area is covered by regular, parallel nano/micro ripples (550 – 750 nm), perpendicular to the laser polarization. At 5 times higher dose (panel b)), the spot center is filled much larger, but shorter macro ripplets (≈ 2.5 – 3 µm), parallel to the polarization, whereas nano/micro ripples, perpendicular to the polarization are still found a the outer edge of the spot. At a very high dose of 1,000 pulses (panel c)), already a crater has formed, on the wall of which pillars of several-µm width are found, still regularly ordered mostly perpendicular to the polarization. The pillars are surrounded by a regular ring of round tip- (or bubble-)like features, appearing almost as a continuation of of the central pillar tips. At the very outer edge, again, nano/micro ripples are formed (cf. Fig. 5b)).

A closer look at the low-dose pattern reveals another interesting feature: In the upper left corner (white arrows in Fig. 4a), two defects (like circular holes) appear to be the centers of circular ripple patterns (panel aa)), of the same feature size as the parallel ripples, but not depending on the laser polarization. Further, even the almost regular ripples are superimposed by a much less regular, perpendicular modulation.

Much more interesting is a closer inspection of the 50-pulses pattern. A magnified detail is shown in Fig. 5 a). Obviously, the macro ripplets are connected by remains of the low-dose ripples. In the upper right corner, such remains are even found as an over-structure on the back of the ripplets. A comparison with the high-dose tip-structure (Fig. 5 b)) suggests that the latter appears as a break-up of the macro ripplets. On the other hand, the fine ripples at the edge are interrupted by a modulation resembling both the low-dose over-structure and the macro ripplets at 5 times higher dose.

4. Discussion

The experimental results indicate that, indeed, there is a continuous EVOLUTION from very fine structures (nano/micro ripples) to increasingly broader and more complex patterns when the irradiation dose is increased. This suggests that the formation of the different patterns is strongly related.This evolutionary correlation is much more in favor of the non-linear structure formation of model B than supporting the involvement of several, DIFFERENT mechanisms in the formation of each structure as, presently, used in model A.

More likely, the experiments point to the following scenario: already at rather low dose, the regular ripples start to undergo a second phase of structuring, resulting in the less regular modulation grossly perpendicular to the original ripples direction. Similar to the point defects (Fig. 4 a), aa)) also the direction of the nano/micro ripples is affected: they start deviating from long straight lines and slightly undulate according to the over-structure. This becomes evident in the magnified detail of the low-dose pattern, shown in Fig. 6. With increasing dose, indeed, this modulation appears to coalesce into macro ripplets, as is indicated in Fig. 6 by the additional display – at the same scale – of the 50-pulses pattern. In the upper right corner, this coalescence is not yet fully reached, and the fine ripples are still visible as a modulation of the ripplet ridge.

This picture is boosted by the observation that the ripplets are interconnected by the remains of the old low-dose pattern. Such interconnecting ladders of fine structures between coarser features is typical for self-organized structure formation, even in completely different fields [27,28]. Also the general phenomenon of evolutionary structure coarsening is a typical feature of non-linear dynamics and is very well reproduced in model (B) [18], whereas it is not at all possible to be considered in the lithographic, passive model (A).

5. Conclusion

We have presented investigations on the evolution of laser-induced surface patterns which strongly support our model of active, self-organized structure formation during relaxation from a laser-induced surface instability. In contrast, the evolutionary correlation cannot be explained by the standard lithographic model for LIPSS formation [2], even not with its recent modifications [21].

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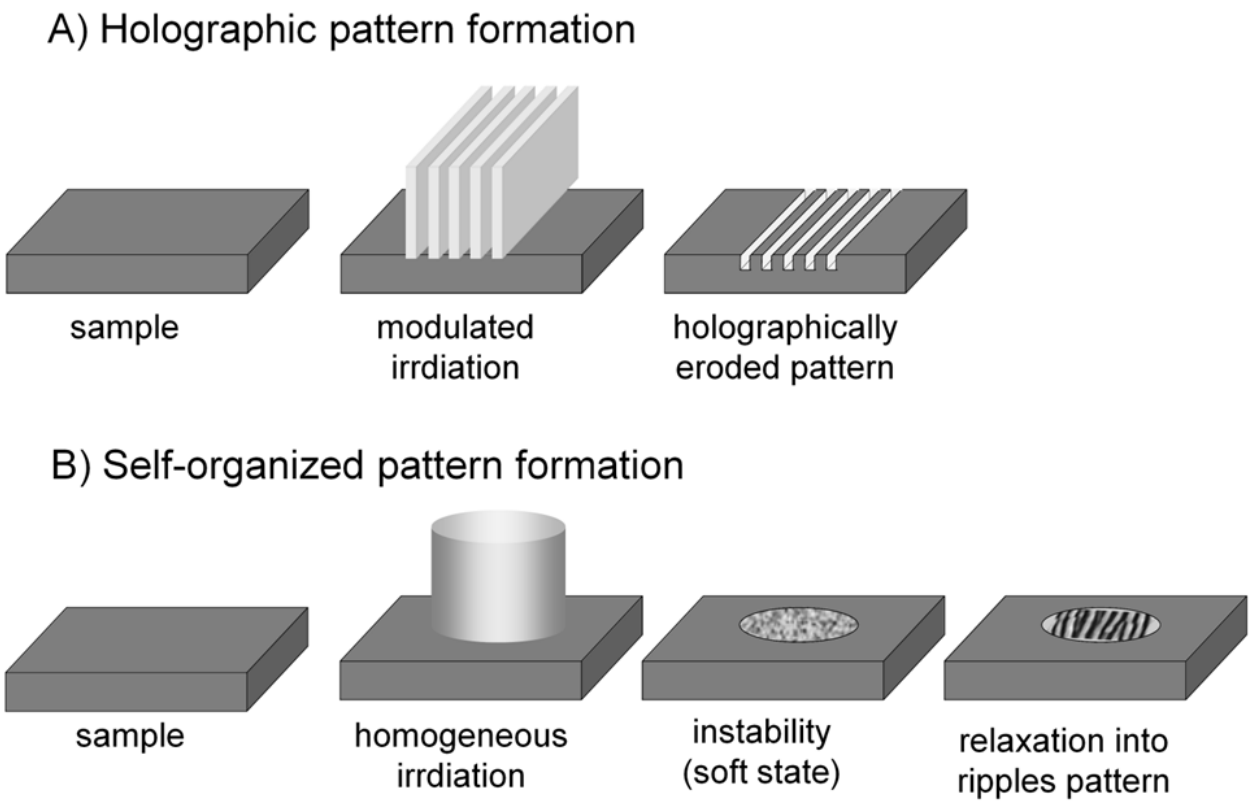

Fig. 1. Two different approaches for laser-induced surface structure formation:
A) modulated ablation by spatially modulated energy input ("interference")
B) self-organized structure formation from a laser-induced surface instability

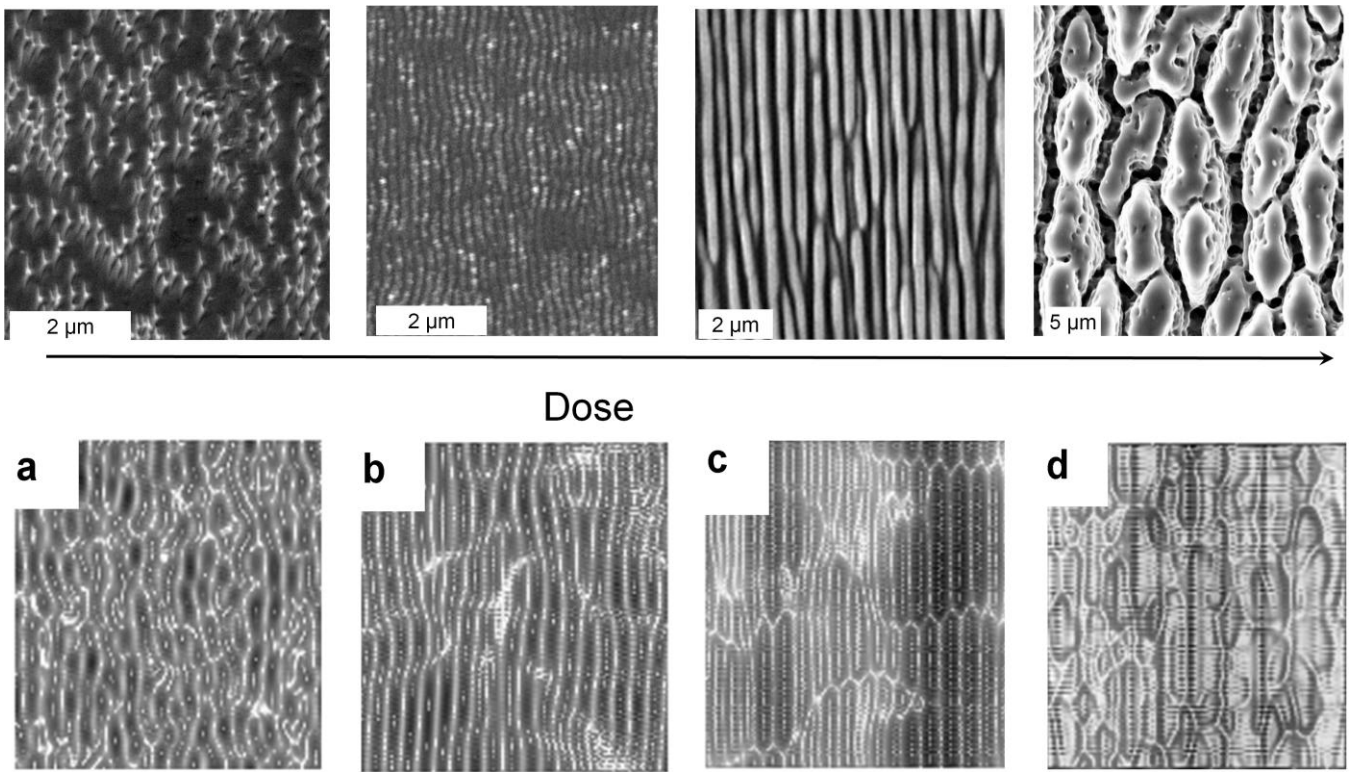

Fig. 2. Changing of laser-induced surface patterns with increasing irradiation dose. Upper row: experimental observation (SEM micrographs); lower row: numerical simulation of self-organized pattern formation [18] (note that for the simulation NO REALISTIC scale can be given, detailed material parameters missing).

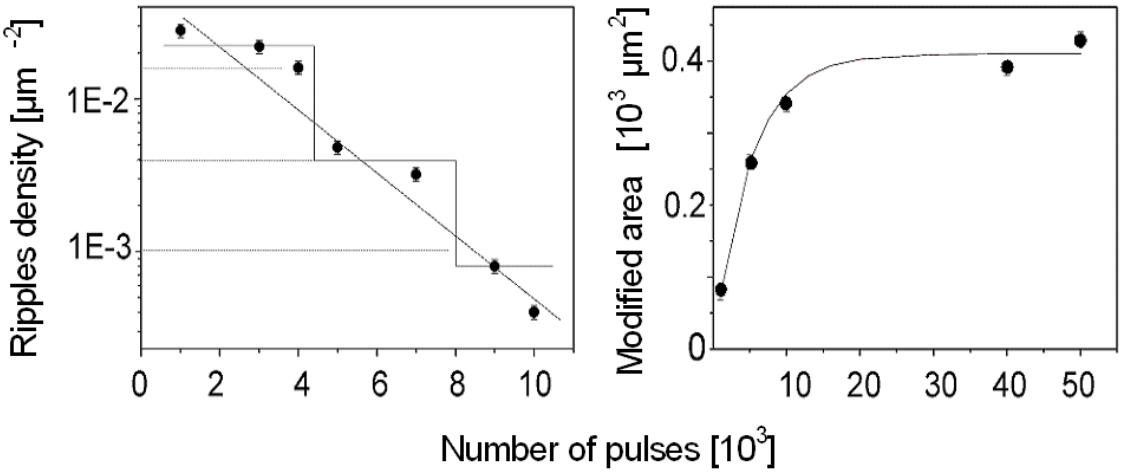

Fig. 3. Dependence of macro-ripples (cf. Fig. 2d) density on silicon (the square root corresponds to the inverse of average feature width Λ) and modified spot area as a function of irradiation dose (number of pulses at 2.6×10^{12} W/cm^2). Note, in the left panel, that the increase in feature size can, not only, be fitted by a simple log-function (straight diagonal line) but also by a discontinuous step distribution. The dashed horizontal lines close to the steps correspond to discontinuous period doubling, as is typical in self-organized structure formation [24] (Note that due to the two-dimensional representation they are separated by a factor of 4).

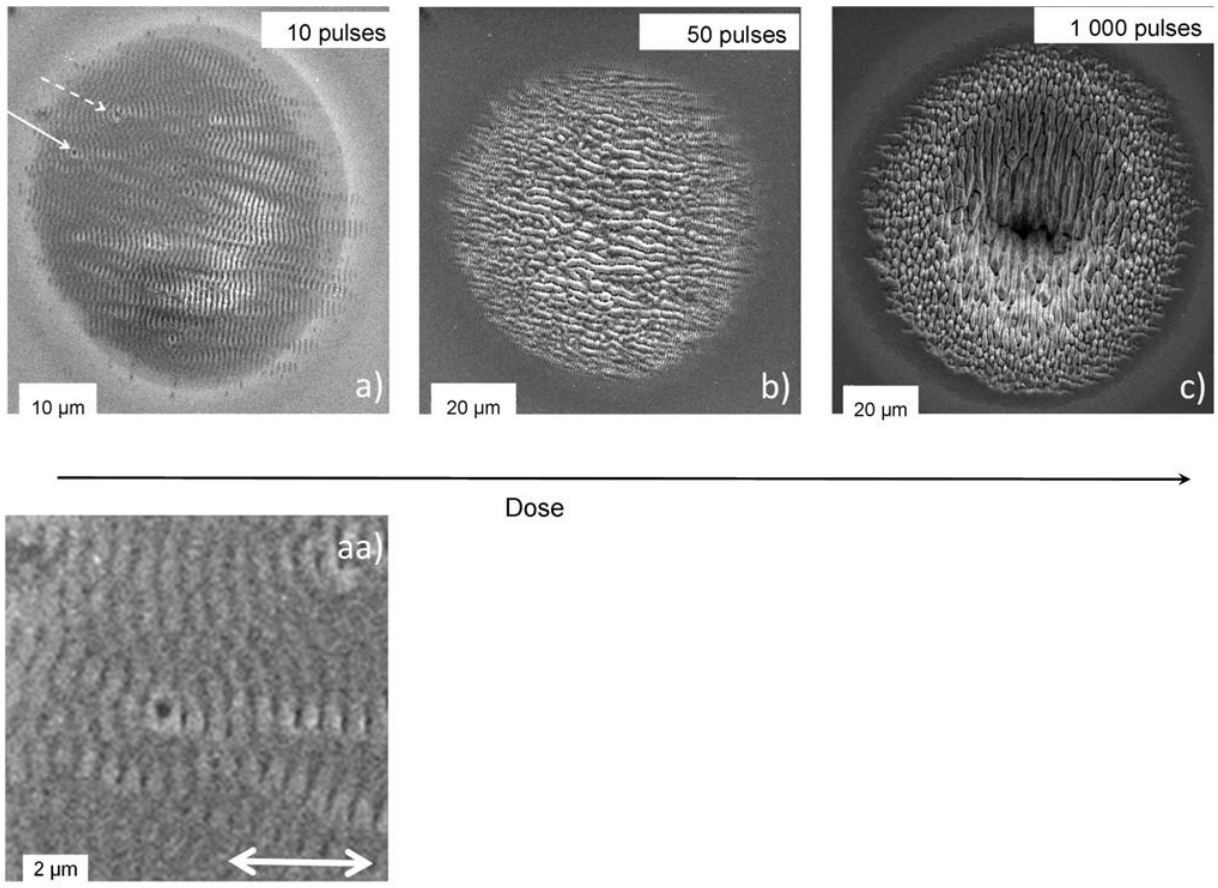

Fig. 4. Evolution of silicon surface morphology with increasing dose (number of pulses at 2.15×10^{12} W/cm^2 [spot center], close to the ablation threshold of $I_{th} = 2\times10^{12}$ W/cm^2 [25,26]). The white arrows in panel a) point to two hole-defects, surrounded by a spherical ring pattern of ripples as shown in the magnification (panel aa)) around the lower whole (solid arrow in a)). The double arrow in aa) indicates the polarization

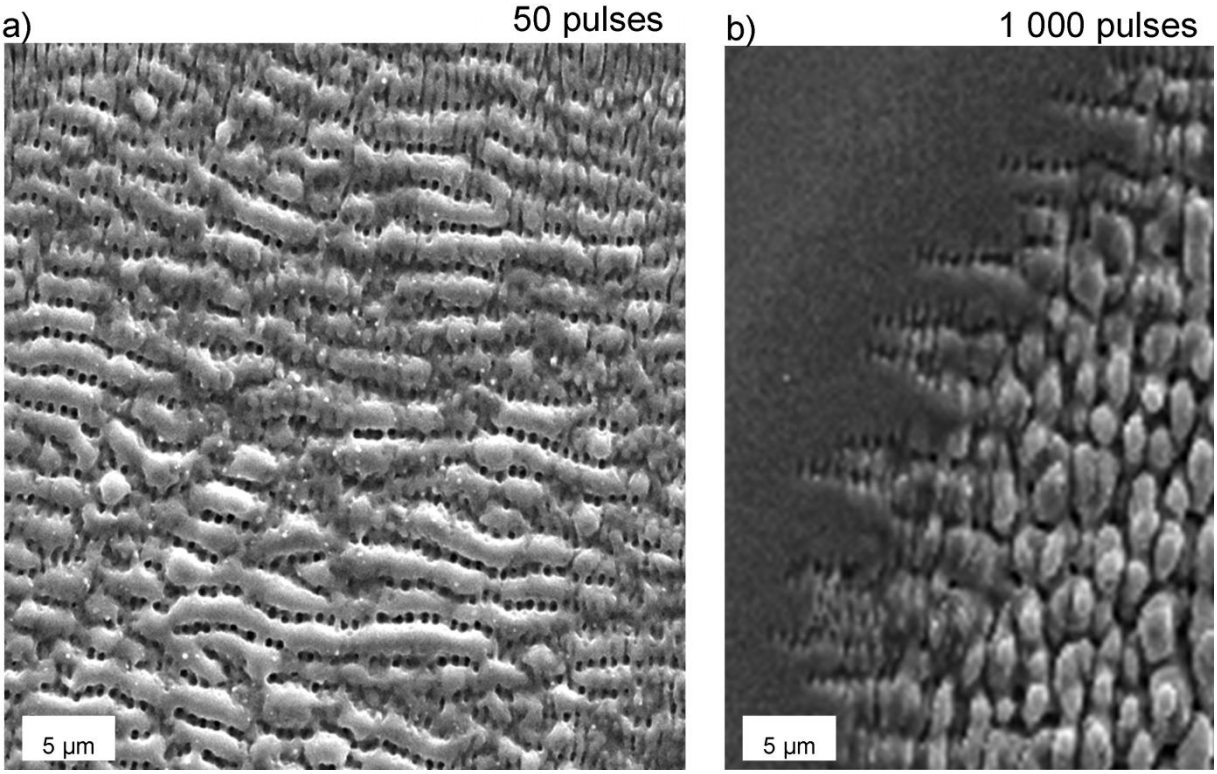

Fig. 5. Details from Fig. 4.
a) central part of the spot at 50 pulses, note the coexistence of fine and super-coarse features (macro-ripples)
b) edge of the spot at 1,000 pulses, note the merging of fine ripples into perpendicular macro- ripples and then the breaking into cones or bubbles..

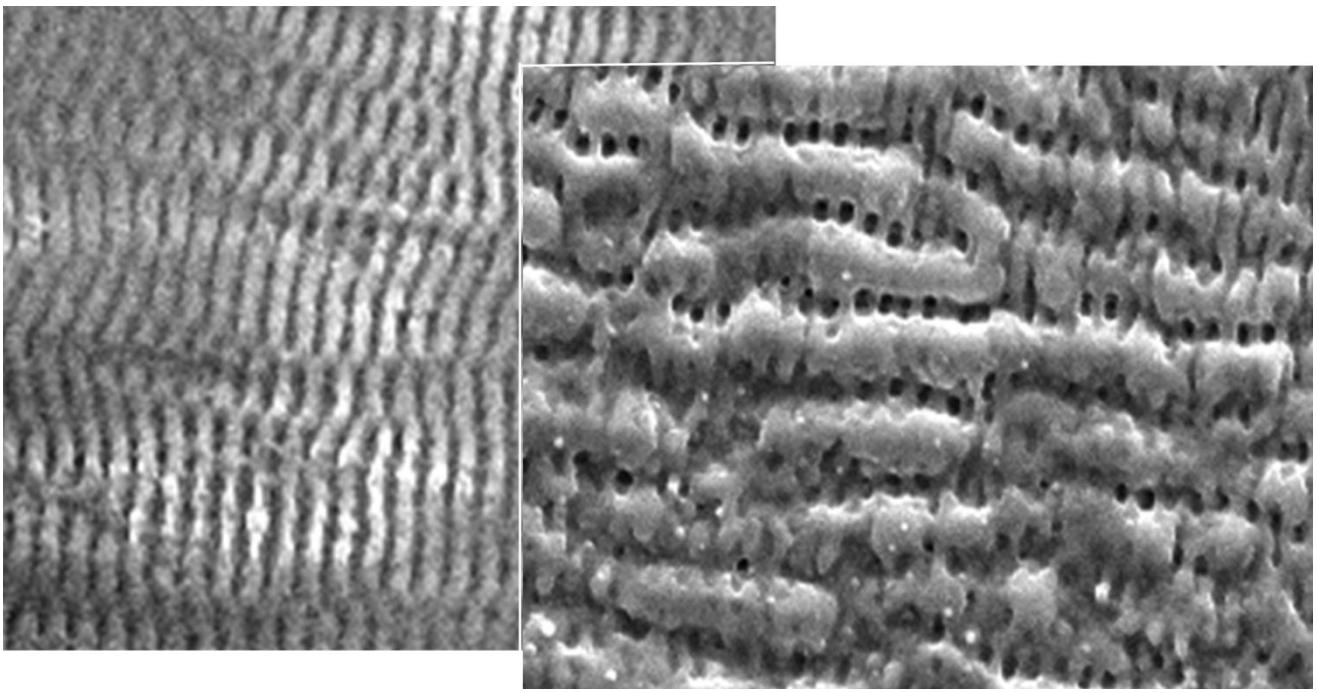

Fig. 6. Details from Fig. 4 (left panel: 10 pulses, right panel 50 pulses; same magnification). Note (left panel) the super-coarse horizontal modulation, perpendicular to the fine ripples, which appears to coalesce into macro- ripples at 5-times higher dose (right panel). In the upper right edge of the right panel, intermediate, already partly coalesced structures are visible.