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Toward a Phenomenological Model of Laser-Driven Patterning of Solid Deformable Substances

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14. ABSTRACT

In this report we suggest an approach to the laser-driven patterning of solids, based on the concepts of Stress-Driven Rearrangement Instabilities and the Phenomenological Mechanochemistry of Damage. We deliberately ignore all the thermal effects in order to make the role of stresses more transparent. If the suggested approach demonstrates its vitality, it should be combined with more traditional approaches. The suggested approach appears to be technically meaningful even in the absence of deformations and stresses. In this approximation, the formulated 1-D boundary value problem allows the exact solution.

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1. Introduction

Laser irradiations lead to various morphological instabilities and surface patterning.[1–6] Some of them are mostly of the macroscopic scale, like those reviewed in Phipps[1] and Allmen,[2] while others show up at micro- and nanoscales.[2–7]

Macroscale instabilities are usually analyzed on the basis of the Stefan problem related to the phenomena associated with the phase transformations liquid–vapor, solid–gas, or solid–liquid.

In this report, we direct the readers' attention to another mechanism of morphological instability, also related to the phase transformation liquid–melt or liquid–vapor. In the 1990s the instability in question was coined the Stress-Driven Rearrangement Instability (SDRI).[8–11] SDRIs essentially rely on two central cornerstones.

The first cornerstone is the ability of substances under study to accumulate internal energy in the form of the standard elastic energy. Thus, we must assume the deformability of the body. Typically, the elastic deformations are very small. They are not able to change significantly the original shapes of the bodies unless they are extremely thin. In those bodies, various stress-driven instabilities show up. The typical instability of this sort is the Euler instability of axially stressed elastic beams as well as instabilities of elastic plates and shells. The Euler instabilities play a crucial role in science and engineering, but they have nothing in common with the SDR instabilities.

The second cornerstone of the SDRI concerns the ability to *rearrange* the body's elements. We define this rearrangement as the ability to change the neighbors of the constituent material's particles. For the laser community, the best example of the rearrangement would be the ablation-like phenomena. In the ablation processes, the shapes of the bodies undergo changes due to elastic deformation and thermal expansion. But the most significant changes of the shape result from the loss of material elements due to the melting–crystallization or vaporization–sublimation processes, both examples of what we have coined as "rearrangement". In the Appendix we provide a more detailed explanation of the mechanism of the SDRIs.

Quite often, the rearrangement is triggered by sufficiently high macroscopic stresses and temperature gradients. In Grinfeld[8–10] and Nozieres[11] we presented theoretical models and instruments allowing analysis of the rearrangements in solids using only macroscopic notions. At the same time, it seems feasible to trigger the ablation by direct influence of radiation on the micro- and nanoscale levels without creating any macroscopic fields within the whole body. In principle, these phenomena can be studied without any macroscopic (homogenized) notions. In Grinfeld[12] and references therein, we suggested an intermediate approach we called the Phenomenological Mechanochemistry of Damage (PMD), combining the macroscopic notions and methods (e.g., stresses and thermodynamics) with nanoscale ones (bonds). The mathematical models of the PMD are considerably simpler than the models of SDRIs and at the same time allow a description of the phenomena, similar to those of the SDRIs, in a much simpler way.

We believe that the SDRI and PMD techniques, and the relevant theoretical and numerical techniques, can be helpful for analysis of laser drilling and for describing surface patterns appearing under the action of laser radiation. In this report we suggest theoretical amendments to the theory described in Grinfeld[9,10,12] and Nozieres[11] that are necessary for addressing those problems.

Theoretical analysis of the SDRI and even PMD involves highly nonlinear equations allowing analytical methods only in the initial stage of unstable growth. Currently we are unable to explore analytically the most-important deeply nonlinear regimes of growth. To avoid this difficulty, researchers develop numerical tools facilitating the process of solving and interpreting the results by means of visualization of developing morphologies.

We took the liberty to ignore all thermal notions. It is not because we underestimate the role of the thermal variables; unquestionably, high temperatures are key in the theory of ablation. We do this only to demonstrate more clearly the potential role of the stresses and rearrangement taken alone.

2. Master System for Analysis of the Laser-Driven Fracture

Per the approach of the PMD, we consider the elastic energy density e per unit mass, which depends upon the displacement gradients $\partial_j u_i$ and the fraction κ of the broken bonds, where $u_i(x,t)$ is the field of the displacements and $\kappa(x,t)$ is the ratio of broken bonds to the total number of bonds; thus, the variable $\kappa(x,t)$ changes with the limits amount $0 \le \kappa(x,t) \le 1$. So, we postulate that

$$e = e(\partial_j u_i, \kappa). \tag{1}$$

The stress tensor p^{ij} and the chemical potential χ with respect to the birth/death of the bonds are defined as

$$p^{ij} = \frac{\partial e}{\partial\left(\partial_j u_i\right)}, \quad \chi = \frac{\partial e}{\partial \kappa}, \tag{2}$$

where mass density = 1.

Now we can formulate the master system. First we assume the inertia can be neglected. Thus, we arrive at the bulk equilibrium equation

$$\frac{\partial p^{ij}}{\partial x^j} = 0. \tag{3}$$

We neglect the mechanical traction at the surface Σ exposed to the radiation. Thus we arrive at the boundary condition

$$p^{ij} n_j = 0, \tag{4}$$

where n_j is the normal to the boundary Σ.

Equations 3 and 4 comprise the standard static system of the mechanics of continua. However, we are dealing not with mechanics but with mechanochemistry. Therefore, we need one more equation for the "chemical" unknown κ.

$$\frac{\partial \kappa(x,t)}{\partial t} = -K\chi + f_{ext}(x,t), \tag{5}$$

where the positive kinetic constant K defines the "interior" kinetics of the defects, and last term, $f_{ext}(x,t)$, defines the bulk production of the damaged bonds due to the "exterior" radiation.

The specifics of the boundary conditions, when dealing with the rearrangement, consist in the fact that the position of the boundary Σ is one of the unknowns [in addition to the standard unknowns $u_i(x,t)$ and $\kappa(x,t)$]. Thus, for mathematical (and physical) consistency of the model we need the appropriate boundary condition on the transient boundary S (in addition to the standard condition [Eq. 4]). This additional condition can be postulated based on different principles. When dealing with ablation it makes sense to assume that the material particles leave the body, exposed to the laser radiation, when the fraction of the broken bonds κ attains certain critical value κ_{crit}:

$$\kappa(x,t)\big|_S = \kappa_{crit}. \tag{6}$$

To get a formally closed mathematical system, we have to choose the functions $e = e\left(\partial_j u_i, \kappa\right)$ and $f_{ext}(x,t)$. In the following, we illustrate reasonable choices of these functions for the 1-D case.

Consider a solid damageable half-space, exposed to radiation, as presented in Fig. 1. The field of displacements has only one component, $u(x,t)$, collinear with the x-axis. We postulate the energy density in the form

$$e(u_x,\kappa)=\frac{1}{2}(1-\kappa)G_0u_x^2+B(\kappa-\kappa_0)u_x+\frac{1}{2}\chi(\kappa-\kappa_0)^2, \tag{7}$$

where G_0, B, χ, and κ_0 are positive constants.

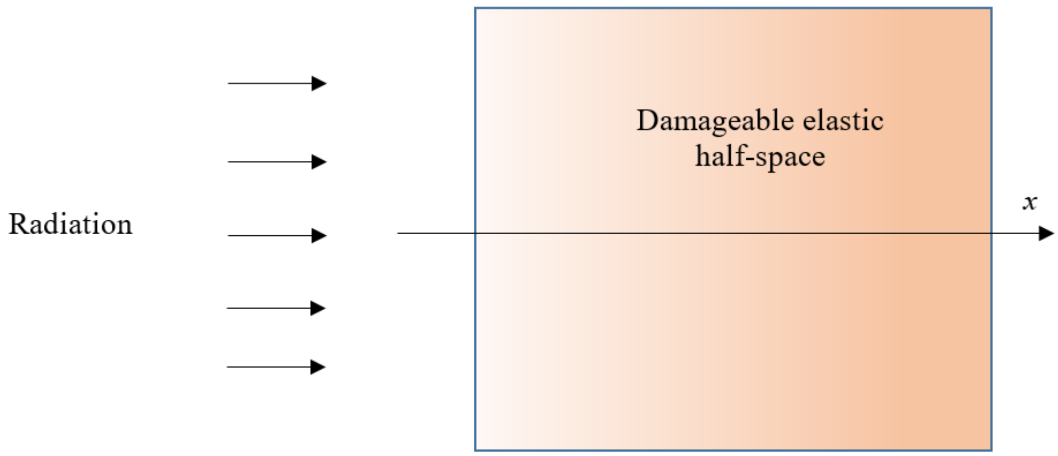

Fig. 1 Damageable elastic half-space exposed to radiation

We then get, differentiating Eq. 7,

$$\begin{aligned}\chi&=e_\kappa(u_x,\kappa)=-\frac{1}{2}G_0u_x^2+Bu_x+\chi(\kappa-\kappa_0)\\ p&=e_{u_x}(u_x,\kappa)=(1-\kappa)G_0u_x+B(\kappa-\kappa_0)\end{aligned}. \tag{8}$$

Using Eq. 8, we can rewrite Eqs. 3 and 5 as follows:

$$\frac{\partial}{\partial x}\left((1-\kappa)G_0u_x+B(\kappa-\kappa_0)\right)=0 \tag{9}$$

and

$$\frac{\partial\kappa}{\partial t}=K\left[\frac{1}{2}G_0u_x^2-\chi(\kappa-\kappa_0)\right]+f(x,t). \tag{10}$$

The traction-free boundary condition (Eq. 4) implies

$$\left[(1-\kappa)G_0u_x+B(\kappa-\kappa_0)\right]_{x=x_S(t)}=0, \tag{11}$$

where $x = x_S(t)$ is the equation of the front.

Combining Eqs. 9 and 11, we arrive at the following solution:

$$u_x(x,t) = -\frac{B}{G_0}\frac{\kappa-\kappa_0}{1-\kappa}, \quad x_S(t) \le x < \infty . \tag{12}$$

Using Eq. 12, we can rewrite Eq. 10 as follows:

$$\frac{\partial\kappa}{\partial t} = K\left[\frac{1}{2}\frac{B^2}{G_0}\frac{(\kappa-\kappa_0)^2}{(1-\kappa)^2} - \chi(\kappa-\kappa_0)\right] + f(x,t). \tag{13}$$

To proceed further we have to choose the exterior flux, $f(x,t)$:

$$f(x,t) = Ae^{-\Delta(x-x_S(t))} . \tag{14}$$

Then we can rewrite Eq. 13 as follows:

$$\frac{\partial\kappa}{\partial t} = K\left[\frac{1}{2}\frac{B^2}{G_0}\frac{(\kappa-\kappa_0)^2}{(1-\kappa)^2} - \chi(\kappa-\kappa_0)\right] + Ae^{-\Delta(x-x_S(t))}. \tag{15}$$

Eq. 15 should be amended with the initial data

$$\kappa(x,0) = \kappa_{in}(x) \tag{16}$$

and the boundary condition

$$\kappa(x_S(t),t) = \kappa_{crit} . \tag{17}$$

For the case where the damage $\kappa(x,t)$ forever remains less than κ_{crit}, the master system (Eqs. 16 and 17) becomes considerably simpler precisely because the movable boundary S remains fixed, so that $x_S(t) = 0$. Then Eq. 15 reads as

$$\frac{\partial\kappa}{\partial t} = K\left[\frac{1}{2}\frac{B^2}{G_0}\frac{(\kappa-\kappa_0)^2}{(1-\kappa)^2} - \chi(\kappa-\kappa_0)\right] + Ae^{-\Delta x} . \tag{18}$$

If $B = 0$, $\kappa(x,0) = 0$ and the system (Eqs. 16 and 18) has the following solution:

$$\kappa(x,t) = \left(1-e^{-K\chi t}\right)\left(\kappa_0 + \frac{A}{K\chi}e^{-\Delta x}\right). \tag{19}$$

For the stationary (i.e., time-independent) solution, Eq. 18 implies

$$\frac{1}{2}\frac{B^2}{G_0}\frac{\left(\kappa-\kappa_0\right)^2}{\left(1-\kappa\right)^2}-\chi\left(\kappa-\kappa_0\right)+\frac{A}{K}e^{-\Delta x}=0\,. \tag{20}$$

3. Negligible Elasticity Effects

The problem of laser-induced ablative drilling, as it is understood in this report, remains meaningful even in the absence of elastic effects. Formally, we can just use Eqs. 9 and 10, assuming $G_0 = B = 0$. In this case, Eq. 9 appears to be redundant, whereas Eq. 10 reduces to

$$\frac{\partial\kappa}{\partial t}=-K\chi\left(\kappa-\kappa_0\right)+f\left(x,t\right). \tag{21}$$

Choosing the flux $f\left(x,t\right)$ in the form (Eq. 14), we arrive at the following equation:

$$\frac{\partial\kappa}{\partial t}=-K\chi\left(\kappa-\kappa_0\right)+Ae^{-\Delta\left(x-x_S(t)\right)}, \tag{22}$$

which should be amended with the initial condition (Eq. 16) and boundary condition (Eq. 17).

The problem (Eqs. 16, 17, and 22) allows for the exact solution, which we describe next without a detailed derivation. The exact solution is particularly simple when the amplitude A is sufficiently small. In this case, the damage distribution $\kappa\left(x,t\right)$ stays below κ_{crit}, the front undergoing radiation remains fixed [i.e., $x_S\left(t\right)=0$], and there is no need for the boundary condition (Eq. 17). Then the exact solution for the $\kappa\left(x,t\right)$ reads as

$$\kappa\left(x,t\right)-\kappa_0=\left(\kappa_{ini}\left(x\right)-\kappa_0\right)e^{-K\chi t}+\frac{A}{K\chi}e^{-x\Delta}\left(1-e^{-K\chi t}\right). \tag{23}$$

At $t\to\infty$, the relationship (Eq. 23) implies

$$\underset{t\to\infty}{\kappa\left(x,t\right)-\kappa_0\to}\frac{A}{K\chi}e^{-x\Delta}\;. \tag{24}$$

Thus, the solutions with an immobile front (i.e., fixed boundary) are possible if and only if

$$\frac{A}{K\chi} \le \kappa_{crit} - \kappa_0 . \quad (25)$$

Otherwise, the relationship (Eq. 23) provides a meaningful solution only up to the moment $t = t_c$, such that

$$\kappa_{crit} - \kappa_0 - \frac{A}{K\chi} = \left(\kappa_{ini}(0) - \kappa_0 - \frac{A}{K\chi} \right) e^{-K\chi t_c} . \quad (26)$$

For instance, if $\kappa_0 = \kappa_{ini} = 0$, the relationship (Eq. 26) reads as

$$1 - \frac{K\chi}{A}\kappa_{crit} = e^{-K\chi t_c}, \quad t_c = \frac{1}{K\chi}\ln\frac{A}{A - K\chi\kappa_{crit}} 0 < t < \infty . \quad (27)$$

At this moment, when κ_{crit} is fixed, the distribution $\kappa(x, t_c)$ is equal to $\kappa_{sw}(x) \equiv \kappa(x, t_c)$:

$$\kappa_{sw}(x) = \frac{A}{K\chi} e^{-\Delta x} \left(1 - e^{-K\chi t_c}\right) = e^{-\Delta x}\kappa_{crit} . \quad (28)$$

At $t > t_c$ the front, undergoing radiation, begins moving and reaches the point $x > 0$ at the moment $t_S(x) > t_c$ such that

$$t_S(x) = \frac{1}{K\chi}\ln\left(\frac{A}{A - K\chi\kappa_{crit}} + \frac{K\chi\kappa_{crit}}{A}\Delta x \right). \quad (29)$$

4. Conclusion

In this report we suggested an approach to the laser-driven patterning of solids based on the concepts SDRI and PMD. In our approach we deliberately ignore all the thermal effects in order to make the role of stresses more transparent. If the suggested approach demonstrates its vitality, it should be combined with more-traditional approaches.

The suggested approach appears to be technically meaningful even in the absence of deformations and stresses. In this approximation, the formulated 1-D boundary value problem allows the exact solution, described by the relationships Eqs. 21–29.

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Appendix. Energy Mechanism of the Stress-Driven Instabilities

It was demonstrated, on general thermodynamic grounds[1–5], that a flat boundary of nonhydrostatically stressed elastic solids is always (morphologically) unstable with respect to "mass rearrangement". The rearrangement can occur via melting–freezing or vaporization–sublimation processes at liquid–solid or vapor–solid phase boundaries, surface diffusion of particles along free or interface boundaries, adsorption-desorption of atoms in epitaxial crystal growth, and the like.

The Stress-Driven Instability (SDRI) phenomena, although discovered for the phase transformation "crystal-melt", have nothing in common with the instabilities, addressed in the classical Stefan mechanism (SM). For example, SMs are essentially irreversible kinetic phenomena; their analysis unavoidably requires the approach of irreversible thermodynamics. The SDRIs are basically reversible phenomena, although some of their secondary aspects can be irreversible.

The intuitive description of the SDRI is very straightforward, as demonstrated in Fig. A-1.

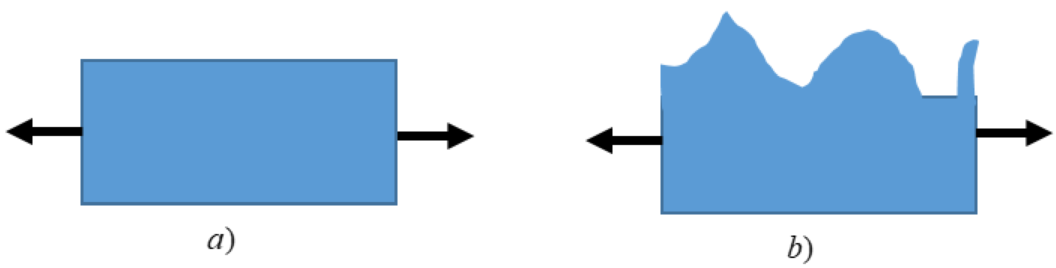

Fig. A-1 Toward the mechanism of the SDRI: a) flat plate uniformly deformed, and b) plate corrugated by action of mass transfer

A flat plate with straight parallel edges is uniformly deformed, as shown in Fig. A-1a). The tractions through the horizontal edges are assumed to be zero, whereas the tractions T through the vertical edges are uniform and do not vanish. Thus there are nonhydrostatic stresses inside the plate with the maximum value $T/2$ (those maximum stresses appear on the cross section having the angle 45° in the horizontal and vertical directions). Let E_{reg} be the total elastic energy, accumulated by this plate with flat edges.

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Assume that the upper stress-free edge of the plate becomes corrugated under the action of mass transfer via one or another mechanism of mass transfer (as shown in Fig. A-1b). Under the action of the same loading at the vertical edges, the plate accumulates another amount of elastic energy, E_{irreg}. If the total masses of the two plates are the same, it can be demonstrated for any model of the elastic substance—nonlinear or linear, anisotropic, or isotropic—that the following inequality is valid:

$$E_{irreg} < E_{reg}. \tag{A-1}$$

The inequality (Eq. A-1) is rather anti-intuitive. The corrugated plate has concentrators of stresses, where the elastic energy density can achieve arbitrary high values. Nonetheless, for the integral energy the inequality (Eq. A-1) appears to be correct.

Obviously, when the mass exchange between the originally flat plate and its melt due to the processes melting–crystallizing is possible, the plate will tend to become corrugated in order to decrease the accumulated elastic energy. In other words, the flat interface is morphologically unstable. Of course, a consistent proof of this fact requires much-more-sophisticated analysis.[2–5]

Many researchers now believe the SDRI is a universal phenomenon over large length scales. This universality allows investigation of nanoscale SDRI effects in, say, semiconductor nanotechnology by means of manifestations of the SDRI in macroscale experiments with He^4 crystals. We discuss the role of SDRIs in the problems of solid nanofilms epitaxy and low-temperature physics. In particular, results related to the dislocation-free Stranski–Krastanov pattern of growth of semiconductor indium-gallium-arsenide quantum dots (QDs) on gallium arsenide substrate grown via molecular beam epitaxy are revealed as an evidence of the SDRI mechanism forming these nanostructures. The structural perfection, epitaxial quality, size, shape and density of the QD and the relationship of these values to the growth parameters were discussed in the 1990s in terms of the SDRI theory. Then, on the basis of the SDRI mechanism, Nozieres[4] and his followers in Europe suggested the qualitative analysis of morphological instabilities of the solid–liquid interface for the macrocrystalline He^4 growing under nonhydrostatic stress.

List of Symbols, Abbreviations, and Acronyms

1-D	one-dimensional
PMD	Phenomenological Mechanochemistry of Damage
QD	quantum dot
SDRI	Stress-Driven Rearrangement Instability
SM	Stefan mechanism

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