Chapter 2

DRY LASER CLEANING OF PARTICLES BY NANOSECOND PULSES: THEORY

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A model for nanosecond dry laser cleaning that treats the substrate and particle expansion on a unified basis is proposed. Formulas for the time-dependent thermal expansion of the substrate, valid for temperature-dependent parameters, are derived. Van der Waals adhesion, substrate and particle elasticity, and particle inertia are taken into account for an arbitrary temporal profile of the laser pulse. The characteristic time for the particle on the surface system is deduced. This time is related to the size of the particles as well as the adhesion and elastic constants. Cleaning proceeds in different regimes if the duration of the laser pulse is much shorter or longer than this time. Expressions for cleaning thresholds are provided and compared with experiments on the 248 nm KrF excimer-laser cleaning of Si surfaces from spherical SiO_2 particles with radii between 235 and 2585 nm in vacuum. Discrepancies between the experimental data and theoretical results seem to indicate that nanosecond dry laser cleaning cannot be explained purely on the basis of one-dimensional thermal expansion mechanism.

Keywords: Dry laser cleaning, modeling, adhesion, threshold, oscillations, SiO_2 particles, Si.

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1. Introduction

Particle removal by means of dry laser cleaning (DLC) (Zapka, 1991; Tam, 1991) has grown in importance during the last decade (Park, 1994; Leiderer, 1998; Heroux, 1996; Bäuerle, 2000). It is used, or consider for usage, in the fabrication of printed circuit boards (PCB), in the production of dynamic random access memory (DRAM) (SIA, 1994; Kern, 1990), in lithography (Teutsch, 1990) and epitaxial growth (Kern, 1993), for the removal of contaminations during via hole production (Lu, 1998a), for the cleaning of

microoptical and micromechanical components. In damage-free DLC expansion of the substrate or particle leads to particle removal. In some cases, other mechanisms related to field enhancement and local ablation may play a role. Another technique is steam laser cleaning (SLC) (Assendel'ft, 1988; Lee, 1991, 1993), which is due to laser-induced explosive vaporization of an auxiliary liquid layer. "Ablative" cleaning is based on the removal of particles/contaminants by ablation. SLC, though more efficient (She, 1999), cannot be applied to hygroscopic materials, and is incompatible with many applications where high purity is required, as in small-scale optics and nanocluster technology. It is also a multiple-step process because of necessary liquid delivery (Halfpenny, 1999).

In this article we concentrate on dry laser cleaning. With DLC it is observed that it is more difficult to remove smaller particles. This has been explained by higher specific adhesion forces. However, accurate theoretical predictions for the dependence of the cleaning fluence on particle size have not been derived. It is not entirely clear which parameters should be optimized to improve the cleaning efficiency and decrease the cleaning threshold, especially with smaller particles. Usually cleaning forces acting on the particles are compared with measured adhesion forces. At the same time, nanosecond laser cleaning takes place over very short time scales, which -- as opposed to conventional adhesion measurements -- requires the consideration of dynamic effects.

Several models of DLC exist. Accelerations and forces due to thermal expansion of the substrate (Tam, 1992; Dobler, 1999) and particles (Lu, 1997a), elastic deformation of the particles which are compressed by the expanding substrate (Lu, 1999), cleaning via generation of surface acoustic waves (Kolomenskii, 1998), the influence of hydrogen bonds (Wu, 1999, 2000), have been considered. The behavior of the particles ***after*** the detachment and the redeposition has been studied (Lu, 2000a; Vereecke, 1999). These models employ many inadequate assumptions. Among these are: Thermal expansion of the substrate and particle are often treated separately and incorrectly. The temporal profile of the laser pulse is not taken into account. This assumes infinite acceleration or deceleration of the substrate in the beginning or end of the laser pulse. Deformation of the substrate and particle, their interdependence and influence of the particle on the substrate expansion are not described properly. Adhesion forces are treated separately from the elastic forces, which can lead to erroneous results. Though the importance of force and energy criteria were mentioned

(Kolomenskii, 1998), their regions of applicability are not clearly stated. The removal of absorbing particles and elasticity of the substrate (Lu, 1996, 1997b) was analyzed on the basis of force balance only (Tam, 1992; Lu, 1997a, 1998b) without taking into account particle movement. The temperatures of particles are estimated in a very crude way there. Dissipative processes are considered in ref. (Lu, 2000a) but only in the post-detachment stage. Numerical calculations do not provide formulas relating cleaning fluence to laser and material parameters.

Our goal is to develop a unified description, which will easily incorporate the influence of different cleaning mechanisms and experimental parameters, and to estimate factors that contribute to DLC, without long numerical calculations. Sound related effects that can be important in picosecond DLC (Mosbacher, 2001; Leiderer, 2000) and SLC (Mosbacher, 1999, 2000), as well as field enhancement effects (Mosbacher, 2001; Lu, 2000b; Luk'yanchuk, 2000, 2001, 2002, Zheng, 2001) are not considered.

2. Adhesion potential and equation of motion

Various forces are responsible for the adherence of particles to a substrate (Mittal, 1988, 1995; Visser, 1976). This section introduces an approximation that takes into account the Van der Waals (VdW) and elastic forces.

2.1. Model expression for VdW-elastic potential

We describe particle-substrate VdW interaction by the energy per unit area j (work of adhesion). It can be obtained by the integration of a Lennard-Jones-like potential acting between two plane surfaces (Muller, 1983). It is related to the Hamaker constant H and Lifshitz-VdW constant V by

$$j = \frac{H}{16\pi e^2} = \frac{3V}{64\pi^2 e^2}. \qquad (1)$$

Here e is the equilibrium distance for the force f or energy u (per area) between two planes:

$$f(z)=\frac{8j}{3e}\left(\left(\frac{e}{z}\right)^9-\left(\frac{e}{z}\right)^3\right),\quad u(z)=\frac{j}{3}\left(\left(\frac{e}{z}\right)^8-4\left(\frac{e}{z}\right)^2\right). \qquad (2)$$

All macroscopic results depend only on j which can be inferred from measurements of the pullout force. We now describe a simple approximation, which we will subsequently employ, and discuss its applicability. If the centers of two spheres (or a sphere and a plane) are moved together by a distance h (see Fig. 1), the energy of the system is the sum of the adhesion energy and the elastic energy. The former can be approximated by

$$U_a=-pa^2j\approx-2prhj\ . \qquad (3)$$

Here r is the particle radius (reduced radius for the case of two particles) and a the contact radius. It is estimated *from geometrical considerations* as $a^2 \gg 2rh$, and we assume that everywhere in the contact region the interaction energy per unit area is $-j$. We estimate the elastic energy from the Hertz contact problem of elasticity theory (Landau, 1986a)

$$U_e=\frac{2\bar{Y}}{5}r^{1/2}h^{5/2},\qquad \frac{1}{\bar{Y}}=\frac{3}{4}\left(\frac{1-s_p^{\,2}}{Y_p}+\frac{1-s_s^{\,2}}{Y_s}\right). \qquad (4)$$

Here, the effective Young's modulus $\bar{Y}$ characterizes the elastic properties of the particle and the substrate. Its value is dominated by the properties of the softer material, where most of the energy is stored. Adding both energies we get the potential and the force acting on a particle in the positive h direction, towards the substrate.

$$U=-2prhj+\frac{2\bar{Y}}{5}r^{1/2}h^{5/2},\quad F=-\frac{\partial U}{\partial h}=2prj-\bar{Y}r^{1/2}h^{3/2}. \qquad (5)$$

This approximation yields equilibrium values of h_0, a_0 and U_0:

$$h_0=\left(2p\,j/\bar{Y}\right)^{2/3}r^{1/3},\quad a_0=\left(2h_0r\right)^{1/2}=\left(2^{5/2}p\,j/\bar{Y}\right)^{1/3}r^{2/3}, \qquad (6)$$

$$U_0=-\frac{3}{5}\left(2pj\right)^{5/3}\bar{Y}^{-2/3}r^{4/3}. \qquad (7)$$

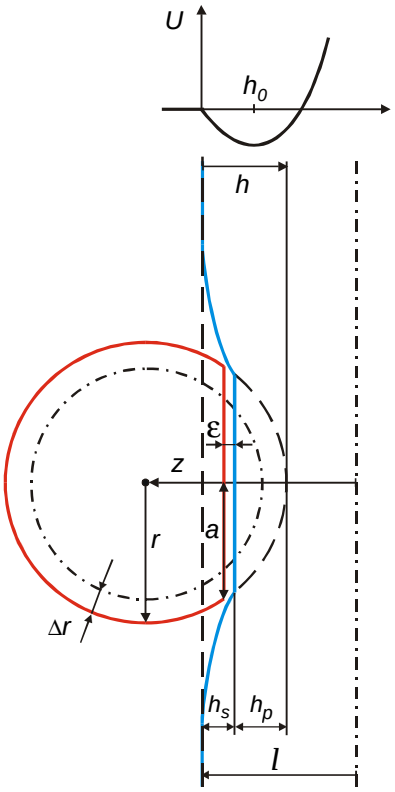

Fig. 1. Schematic of the particle-substrate deformation at a given moment of time. Solid lines - current boundaries of substrate and particles. Dashed lines - their imaginary non-deformed boundaries. Dash-dotted lines - initial position of the substrate and the surface of non-heated (but displaced) particle. l - surface displacement of the substrate, r - radius of the heated particle, Dr - particle expansion, z - position of the particle center referred to initial substrate surface without particle (note, that in general substrate is deformed even before the expansion). Overall deformation is characterized by the so-called approach h; h_s and h_p are its fractions that belong to the substrate and the particle. a - contact radius. e - equilibrium distance between (plane) adhering surfaces. Arrows indicate positive directions for the corresponding quantities. Adhesion potential $U = U(h)$ is also schematically shown. For the depicted moment h is bigger than the equilibrium deformation h_0 (compression stage).

The *maximum* (pull-out, detachment) force is achieved at $h = 0$ and is given by

$$F_0 = 2prj \ . \tag{8}$$

The real situation is more complex and has been studied for a long time (Hertz, 1881, Bradley, 1932). The contact area in the Hertz problem is twice smaller than the geometrical area, $a^2 = hr$. On the other hand, attractive forces near the edges and outside the contact area increase a, modify the profile of the Hertzian gap, and change elastic deformation. Thus, one has to calculate the problem self-consistently (Muller, 1983; Maugis, 1992). Relatively compact results (Muller, 1983; Maugis, 1992; Greenwood, 1998) as well as the earlier JKR (Johnson, Kendall, Roberts) model (Johnson, 1971) predict the following: The contact area is bigger than the Hertzian; tensile stresses exist near the edges; detachment occurs abruptly (with finite contact area) at negative h and has an (small) energy barrier. Nevertheless, these models and expression (5) are similar. A comparison of calculated potentials is shown in Fig. 2. For somewhat different DMT (Derjaguin, Muller, Toropov) model (Muller, 1983; Derjaguin, 1975) the agreement is comparable. The following features can be seen:

- The elastic energy with strong deformation is conveyed correctly, as in this region the adhesion energy is negligible.
- The adhesion part of the energy and pull-out force are linear in radius. Expression (8) for F_0 coincides with the famous Bradley (Bradley, 1932) and DMT (Muller, 1983) results and is 4/3 times bigger than in the JKR (Johnson, 1971) limit.
- The functional dependences for important parameters are correct.
- The total adhesion energy and the difference between the equilibrium h_0 and h at detachment are quite similar. The potential (5) approximates the exact potential shifted by the value of h at the detachment point.
- The contact radius $a(h)$ is conveyed less accurately. For this reason, we avoid using it and express everything via the approach h.

The expressions (5)-(8) should be considered as approximations valid within 20-25%. One can also use the measured pull-out force F_0 and the separation approach h_0 as main parameters of potential. In this form, results apply also to non-spherical particles. The work of adhesion j is actually always calculated from the measured value of F_0. All measurable quantities can be expressed in terms of h_0 and j (or F_0, or U_0, or w_0 introduced below).

2.2. Parabolic approximation

The potential (5) is a smooth function as it is determined mainly by macroscopic elasticity. We can linearize the problem, approximating ***U(h)*** by the parabolic well of the same depth with the minimum at h_0.

$$U \approx U_0\left(1-\left(\frac{h-h_0}{h_0}\right)^2\right). \tag{9}$$

Oscillations within this potential have frequency and period:

$$w_0 = \sqrt{\frac{2|U_0|}{mh_0^2}} = \frac{3}{5^{1/2}(2p)^{1/3}}\left(\frac{\bar{Y}^2 j}{r_p^3 r^7}\right)^{1/6} \sim \left(\frac{v_0^4 j}{r r^7}\right)^{1/6} \sim 0.5\times 10^9 s^{-1}, \tag{10}$$

$$t_0 = 2p/w_0 \approx 10 \text{ ns}, \tag{11}$$

where m is mass of the particle. The estimations assume $r \sim 1$ μm and the typical values listed in Table 2. Note that the sound velocity $v_0 \sim (\bar{Y}/r)^{1/2}$. The approximation based on the second derivative of (JKR or DMT) potential near the equilibrium point leads to a similar frequency, as can be inferred from Fig. 2.

This frequency is ***not related*** to sound vibrations as the particle is treated quasi-statically. This is allowed as long as w_0 is much smaller than the frequency of the first mode of particle sound vibrations. For the estimations we use the expression (Landau, 1986b).

$$w_0 << w_{sound} \approx \frac{p}{2}\frac{v_{0p}}{r} \Rightarrow \frac{6}{p\,5^{1/2}(2p)^{1/3}}\left(\frac{j\bar{Y}^2}{r r_p^3 v_{0p}^6}\right)^{1/6} << 1\,, \tag{12}$$

$$\text{which yields} \quad r >> \frac{1}{2^6}\frac{j}{\bar{Y}} \sim 1.6\times 10^{-12}\text{cm}.$$

This is always fulfilled. As $w_{sound} \mu r^{-1}$ and $w_0 \mu r^{-7/6}$ this condition is almost independent on r.

Many conclusions follow from the linear approximation. The most convenient parameters for the analysis are the pull-out distance from equilibrium, h_0 and the internal frequency w_0. To increase generality, we

also express this frequency in terms of h_0 and measured pull-out force or total adhesion energy. In this form, subsequent results can be applied to other potentials.

$$w_0 = \sqrt{\frac{F_0}{mh_0}} = \sqrt{\frac{2|U_0|}{mh_0^2}}, \quad U_0 = -\frac{F_0 h_0}{2}. \tag{13}$$

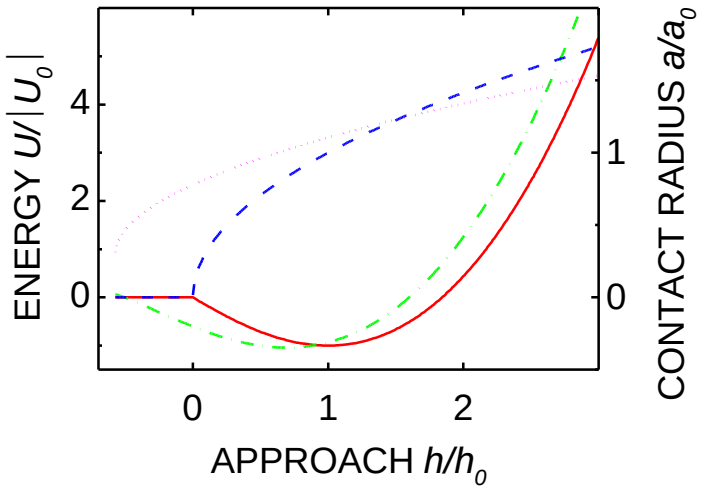

Fig. 2. Exact and approximate elastic-VdW adhesion potential and contact radius in dimensionless variables. Solid line - approximate potential (5). Dash-dotted line - exact potential from the eq. (38) in (Muller, 1983). Dashed line - approximate contact radius (see Eq. (3)). Dotted line - exact contact radius given below the eq. (38) in (Muller, 1983).

2.3. Equation for the evolution of deformation h

When both particle and substrate move, detachment and adhesion are determined by the approach h. Let z be the coordinate of the particle center and l the surface displacement in the *laboratory* frame, both counted from the *initial* position of the substrate surface (see Fig. 1). Then h is given by

$$h = l + r - z. \tag{14}$$

Here all quantities may depend on time: l and r due to the thermal expansion of the substrate and the particle, and z due to particle movement. The force balance should be written for $m\ddot{z}$ (dot stands for time derivative). Rewriting Newton's equation with the force (5) for h instead of z, with the help of (14), we obtain the equation for the evolution of h:

$$\ddot{h} + g\dot{h} = \frac{1}{m}\left(2\pi r j_\infty - \bar{Y} r^{1/2} h^{3/2}\right) + (\ddot{l} + \ddot{h}) \,. \qquad (15)$$

Though the term $m\ddot{l}$ can be interpreted as the force of inertia in the frame moving with the substrate, and the term $m\ddot{h}$ cannot, both the substrate and particle expansion can be treated similarly. All other terms can be neglected in the first approximation.

2.4. Damping coefficient

With the small particles and substantial velocities involved, dissipative processes may become significant. The importance of damping in Eq. (15) is difficult to estimate.

2.4.1. Knudsen viscosity

The motion of the particle is slowed down by the ambient medium. At spatial scales smaller than the mean free path of ambient gas molecules, the damping force can be estimated as:

$$F_d \approx \frac{m_a^2 p}{3(k_B T)^2} \pi r^2 v_T^3 \dot{z} \quad \Rightarrow \quad g \sim \frac{4\sqrt{2}}{p\sqrt{p}} \frac{N\sqrt{m_a k_B T}}{r_p r} \sim \frac{r_g v_0}{r_p r} \overset{gas}{\sim} 10^6 \,\mathrm{s}^{-1} \,. \qquad (16)$$

Here, k_B is the Boltzmann constant, m_a is the mass of gas molecules, v_T their thermal velocity, p the pressure, T the temperature, and N the number density of the gas. The force is defined following the method given in (Lifshitz, 1981).

2.4.2. Stokes viscosity

With bigger particles and / or a (thick) liquid layer at the surface, g can be estimated from the Stokes formula:

$$g = \frac{6phr}{m} = \frac{9}{2}\frac{r_l n}{r_p r^2} \overset{gas}{\sim} \frac{v_0 r_a}{r^2} \sim 10^5 \mathrm{s}^{-1}\,, \tag{17}$$

where h is the dynamic viscosity of the ambient, n the kinematic viscosity and r_a typical molecular size. Both viscous mechanisms, together with thermophoresis, seem to be more important *after* the detachment, in particular for the redeposition problem.

2.4.3. Absorption of sound

The ultrasound generated by the thermal expansion may be strongly damped. As an estimate one can use the rate of energy dissipation in the sound waves (Landau, 1986c)

$$g \approx \frac{w_c^2}{2v_0^2}\left(n + D\frac{bv_0^2}{c}\frac{bDT}{3}\frac{1+s}{1-s}\right). \tag{18}$$

Here D is the thermal diffusivity of the material, c the specific heat, b the thermal expansion coefficient, and w_c some characteristic frequency. The temperature rise $DT \sim T$. With a small particle size, the situation can become even more complicated due to reflection of the sound and temperature wave from the boundaries. In any case,

$$g \sim \max\left(n,\, D\frac{bv_0^2}{c}\, bT\right)\frac{1}{d^2} \sim 10^8\, s^{-1}, \tag{19}$$

where d is some characteristic length, $d \sim r,\, v_0 t,\, v_0 / w_c, \sqrt{Dt}$, depending on the parameters and geometry. The two terms in brackets are related to viscous damping and dissipation by heat conduction due to adiabatic temperature variations within the sound wave. Finally, estimation in Eq. (19) is performed with the typical values from Table 2. It agrees with logarithmic decrements of 10^{-1}-10^{-3} given in (Gray, 1972). In any case expression (19) seems to be smaller than the energy losses due to ***emission*** of sound.

2.4.4. Emission of sound

The particle, which oscillates on the surface with a frequency w_0, *emits* sound into the substrate. This seems to be a primary mechanism of energy loss. Dipole approximation from ref. (Landau, 1987) for sound emission from a sphere oscillating in a liquid yields the following power:

$$P = \frac{2\rho r_s}{3v_0^3} u^2 \frac{r^6 w_0^4}{4 + (w_0 r / v_0)^4}. \tag{20}$$

Here, u is the amplitude of the velocity. The rate of loss for the energy E is given by

$$g \approx P / E \quad \text{with} \quad E = \frac{mu^2}{2} \tag{21}$$

leading to

$$g \approx \frac{r_s}{r_p} \frac{v_0}{r} \frac{1}{1 + 4(v_0 / w_0 r)^4}. \tag{22}$$

This loss can be smaller as the particle is not completely immersed in the substrate material. As a frequency we can use w_0 from Eq. (10). In any case all expressions for the damping coefficient are only estimates.

2.4.5. Plastic deformations

With sub-μm adhering particles, stresses near the edge of the contact area may exceed the tensile strength of the material leading to plastic deformations (Mittal, 1988; Johnson, 1971). In this case, the contact area becomes bigger than that used in the Eq. (3) and below. As a result, the expression for the adhesive force needs to be significantly modified. Another effects can be due to slow formation of covalent bonds between the particle and the surface (Wu, 2000). Phenomenologically, the can be described as an increase in work of adhesion j. With real particles these effects usually become more pronounced with smaller particles, where stresses are larger. There are experimental indications that such effects may

take place not only with soft polymer particles, but even with relatively hard sub-μm colloidal silica particles as well.

3. Thermal expansion

To study ***dry*** laser cleaning the thermal expansion of the substrate and the particle must be calculated. Their independent contributions are combined in Eq. (15). We start with the thermal expansion of the substrate.

3.1. Hierarchy of scales

In the present problem there exists a certain hierarchy of scales. The spot sizes typically employed in laser cleaning are rather big. As a result, the ***axial*** (*z*) extension of the thermal field is much smaller than its ***lateral*** (*x-y*) dimension (laser spot size w_0), even with "weakly absorbing" substrates.

$$l_a + l_T < w_0 . \tag{23}$$

Here l_T is the heat diffusion length (Bäuerle, 2000) and l_a the absorption length. If the sound does not leave the heated region in the ***axial*** direction during the laser pulse, i.e., if

$$v_0 t < (l_a + l_T) < w_0 \tag{24}$$

the ***dynamic*** equations of elasticity must be considered. As long as the sound wave is ***within the laser spot size***

$$v_0 t < w_0 \tag{25}$$

the problem is one-dimensional. That is, thermal expansion is ***dynamic***, but ***unilateral*** and only ***axial*** displacement $u_z \neq 0$ exists. If the sound wave leaves the heated area in *z*-direction, but is still within the lateral extension of the source,

$$(l_a + l_T) < v_0 t < w_0 \tag{26}$$

the expansion is ***quasi-static*** and ***unilateral***. This case is most applicable for dry cleaning with ns laser pulses. With $v_0 \sim 10^6$ cm/s, $a \sim 10^{-4}$ cm^{-1} and $w_0 \sim 1$ cm, this yields 10^{-10} s < t < 10^{-6} s. Here, quasi-static compressive stresses in *x-y* planes influence expansion in *z* direction via Poisson ratio. When sound leaves the irradiated spot in ***lateral*** direction, i.e., with

$$w_0 < v_0 t \tag{27}$$

lateral compression relaxes. The elastic problem is again quasi-static, but three-dimensional (3D). As long as Eq. (23) holds, heat conduction is still 1D, which allows one to obtain general results, ***different*** from the quasi-static unilateral expansion.

Finally, when the heat diffuses out of irradiated spot, i.e., with

$$w_0 < l_T \Rightarrow Dt > w_0^2 \tag{28}$$

heat conduction becomes 3D. The result depends on the laser beam profile, and universal analytical solutions do not exist.

Note that with a ***stationary*** temperature distribution and a ***semi-infinite*** substrate the surface displacement is infinite. At big distances from the source the temperature rise decreases as *1/r*, and the surface displacement $l \propto \int_0^\infty T(z)dz$ diverges. Indeed, a stationary temperature distribution in the semi-infinite substrate requires an infinite time and energy. The stationary surface displacement for CW irradiation will be determined by the size of the specimen, and by the heat exchange with the surrounding. For example in ref. (Welsh, 1988), integral expressions for the surface displacement are given. These integrals, however, logarithmically diverge at any point.

3.2. General equations

The equations of ***classical*** isotropic thermoelasticity (Landau, 1986a; Sokolnikoff, 1956; Parkus, 1976) can be written in the form:

$$\rho \ddot{\mathbf{u}} = \frac{Y(1-s)}{(1+s)(1-2s)} \operatorname{div}\operatorname{grad}\mathbf{u} + \frac{Y}{2(1+s)(1-2s)} \operatorname{rot}\operatorname{rot}\mathbf{u} - \frac{b}{3}\frac{Y}{(1-2s)}\operatorname{grad} T. \tag{29}$$

Here, ρ is the density of the material, $\mathbf{u}$ the displacement vector and T the temperature difference from the ambient temperature. Y is Young modulus,

s the Poisson ratio, and b the coefficient of ***volumetric*** thermal expansion. It is three times larger than the coefficient of ***linear*** thermal expansion.

We employ the heat equation in the simplest form

$$c r \dot{T} = div(K\, grad\, T) + Q\,. \tag{30}$$

Here K is the thermal conductivity, c is the specific heat of the material, and Q is the source term. With very short pulses when dynamic (sound) terms are retained in Eq. (30), the assumption of infinite speed of heat propagation in Eq. (30) is not always valid, and equations (29) and (30) should be modified, (Tamma, 1997; Chandrasekharaiah, 1998). However, ***ns*** laser cleaning can be studied without these details. Likewise omitted are effects of high electronic temperature (Anisimov, 1995), shock wave generation, etc. This is justified by the non-destructive processing conditions required in laser cleaning.

We assume the following boundary conditions for equations (29)-(30). Firstly, there is no force at the free surface:

$$s_{xz} = s_{yz} = s_{zz} = 0 \quad \text{at} \quad z = 0\,, \tag{31}$$

where stress s_{ik} is related to strain via ***generalized*** Hook's law:

$$s_{ik} = \frac{Y}{1+s}\left(u_{ik} + \frac{s}{1-2s}u_{ll}d_{ik}\right) - \frac{b}{3}\frac{Y}{1-2s}Td_{ik}\,,$$
$$u_{ik} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i}\right). \tag{32}$$

Secondly, we assume no heat losses into the ambient

$$\frac{\partial T}{\partial z}\Big|_{z=0} = 0\,. \tag{33}$$

And finally, with semi-infinite substrate all quantities disappear inside the material at $z \to \infty$.

Though general solutions of such a problem are possible (Sokolnikoff, 1956; Parkus, 1976), they are not elucidating. For this reason we will discuss the most relevant cases.

3.3. Unilateral quasi-static expansion

In this case all quantities are independent of *x*, *y* and *t*, and only a *z*-component of displacement $\boldsymbol{u}$ is present. For brevity, we will denote it as u and use index z for differentiation with respect to z. We introduce the coefficient of *unilateral* thermal expansion

$$b_1 = \frac{b}{3}\frac{1+s}{1-s}. \tag{34}$$

The equations and the boundary conditions then reduce to

$$u_{zz} = b_1 T_z \quad , \quad u_z\big|_{z=0} = b_1 T \quad , \quad u, u_z\big|_{z\to\infty} = 0, \tag{35}$$

$$cr\dot{T} = (KT_z)_z - I_z \quad , \quad T_z\big|_{z=0} = 0 \quad , \quad T\big|_{z\to\infty} = 0. \tag{36}$$

Here I is intensity of laser light inside the material. Integrating Eq. (35) over z and using boundary conditions at infinity, we get:

$$u_z = b_1 T. \tag{37}$$

One can immediately write

$$u(0) = -\int_0^\infty b_1 T dz. \tag{38}$$

It is possible to avoid solving the heat equation, even with temperature-dependent parameters. We differentiate Eq. (37) with respect to time and substitute T from Eq. (36):

$$\dot{u}_z = \frac{b_1}{cr}(KT_z - I)_z. \tag{39}$$

Integrating over z and using the boundary conditions, we obtain for the displacement vector at the surface.

$$\dot{u}(0) = -\frac{b_1}{cr} I(0). \tag{40}$$

Finally, we introduce absorptivity A and the *transient* absorbed fluence,

$$f_a(t) = \int_0^t AI(t')dt' . \qquad (41)$$

Then, integrating Eq. (40) over t, we obtain the surface displacement $u(0) < 0$. It is negative, as z was directed into the substrate. For the (positive) surface expansion l and for the expansion velocity that enter Eq. (15) we obtain:

$$l(t) \equiv -u(0) = \frac{1+s}{3(1-s)} \frac{b f_a(t)}{cr} \quad , \quad \dot{l} = \frac{1+s}{3(1-s)} \frac{bI_a(t)}{cr} . \qquad (42)$$

Qualitatively, this result is expected, as both the thermal expansion and the heat content within the material are proportional to the absorbed energy. It can be used even for temperature-dependent parameters. Indeed, the thermal conductivity and absorption coefficient, which change strongly for some materials, do not enter this expression. The absorption coefficient may be non-linear in intensity, etc. With temperature-dependent thermal expansion one has to substitute in the elastic equations, in particular in Eq. (37):

$$b_1 T \to \int_0^T b_1(T')dT' . \qquad (43)$$

However, the time differentiation of Eq. (37) keeps Eq. (39) valid if $b_1(T)$ (and $b(T)$) is a ***differential*** coefficient as defined by the Eq. (43). The ratio b_1/cr is approximately constant due to the Grüneisen relation (Gray, 1972; Ashcroft, 1976; Landau 1980) and the result (42) does not change. Temperature variations in the Poisson ratio and the Young modulus are usually insignificant (Landolt-Börnstein, 1982). As Young modulus does not enter the answer, Eq. (42) probably holds also if Y depends on temperature, as in the case of polymers.

Typical surface displacement, velocity and acceleration for an excimer laser pulse of the form

$$I(t) = I_0 \frac{t}{t} \exp\left(-\frac{t}{t}\right) \qquad (44)$$

are shown in Fig. 3. With this definition, the laser fluence is given by $f = I_0 t$ and the pulse duration at the full widths at half-maximum by $t_{\mathrm{FWHM}} \approx 2.45t$.

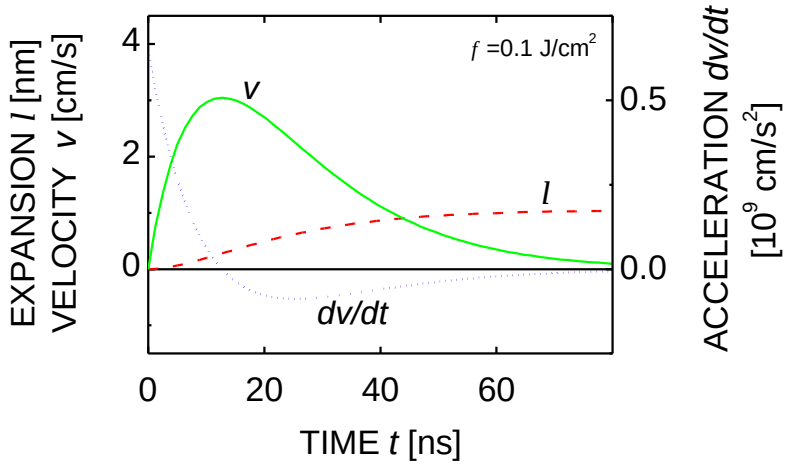

Fig. 3. Surface displacement l (dashed line), velocity v (solid line) and acceleration dv/dt (dotted line) for silicon substrate and typical temporal profile of the laser pulse given by Eq. (44). Laser fluence f = 0.1 J/cm^2. Other parameters used in the calculations are listed in Table 2.

3.4. 3D quasi-static expansion for finite beams with 1D heat conduction

If the condition (27) is fulfilled, the stresses relax outside of the beam area, but the heat conduction can still be considered as 1D. The mathematical derivation is given in the Appendix A. Replacing the integral in Eq. (A.16) by the fluence, as in transition from the Eq. (38) to Eq. (42), we obtain:

$$l(t) = \frac{2(1+s)}{3}\frac{b f_a(t)}{cr} \quad , \qquad \dot{l} = \frac{2(1+s)}{3}\frac{b I_a(t)}{cr} . \tag{45}$$

Although the laser spot size *does not* enter the formula *explicitly*, the consideration *implicitly* assumes that stress and displacement disappear at infinity. This requires 3D relaxation, which can be treated quasi-statically only if Eq. (27) holds.

This result *does not* depend on the spatial profile of the laser beam. Similar results in the particular case of epicentral displacement for the Gaussian beam with surface absorption can be obtained from ref. (Prokhorov, 1990), or found in ref. (Vicanek, 1994).

Comparing Eq. (42) and Eq. (45), we see that the displacement in the latter case is always ***larger*** than the unilateral quasi-static one, as $s < 1/2$. The reason is that lateral stresses partly relax and the material as a whole is "less compressed". Though not all dilatation goes into *z* direction, overall increase in volume still makes the surface displacement bigger than in the unilateral case. The transitional stage $v_0 t \approx w_0$ can be treated only dynamically. In numerical calculations (Spicer, 1996) one can see the unilateral stage and the much slower 3D stress relaxation stage of the expansion.

3.4.1. Comparison between different approximations

The formulas used in the literature for the thermal expansion of the substrate differ in the dimensionless coefficient in expressions such as Eqs. (38), (42) and (45). The results are summarized in Table 1. The difference between various approximations can easily reach a factor of two. Clearly, one should not consider smaller effects, for example related to moderate temperature dependences of parameters, unless correct formula is used.

Table 1. Coefficient of proportionality between the surface displacement and absorbed fluence, that enters (42) and similar quasi-static expressions, and its value for representative values of Poisson coefficient.

Approximation	1D linear expansion	1D expansion	3D expansion 1D heat cond.	1D incomp expansion
Coefficient in Eqs. (42), (45)	$\frac{1}{3}$	$\frac{1+s}{3(1-s)}$	$\frac{2}{3}(1+s)$	1
Poisson ratio s				
-1	1/3	0	0	1
-1/2	1/3	1/9	1/3	1
0	1/3	1/3	2/3	1
1/4	1/3	5/9	5/6	1
1/2 (incomp.)	1/3	1	1	1

3.5. Particle influence on the expansion of the substrate

In some references (Lu, 1997a) for the absorbing particle, (Wu, 2000), for the absorbing substrate) the following expression describes the thermal stress at the surface.

$$s_{zz} \sim YbT \,. \tag{46}$$

This stress exists if the substrate (particle) *is not allowed* to expand. In reality, the expansion is restricted only by the elasticity and inertia of the particle. The latter is not that big due to the small particle size.

Let us consider an expanding substrate. The surface displacement during a nanosecond pulse is of the order of several nanometers (see Eq. (42) and experimental results (Dobler, 1999)). The radius of the particle is at least 10-100 times larger. If the particle does not move, expansion results in an indentation of depth ~ l over an area ~ $a^2 \sim lr$ on the substrate. Such a deformation requires a force ~ $Y_s (lr)^{1/2} l \sim Y_s l^{3/2} r^{1/2}$ (see ref. (Landau, 1986a) or consider elastic part of the force in Eq. (5) with $h \sim l$). Such a force would have resulted in the following acceleration, velocity, and displacement of the particle at the end of the pulse:

$$\ddot{z} \sim \frac{Y_s r^{1/2} l^{3/2}}{r_p r^3} \sim v_0^2 \frac{l^{3/2}}{r^{5/2}}\,, \quad \dot{z} \sim \ddot{z} t \sim v_0^2 \frac{l^{3/2}}{r^{5/2}} t\,, \quad z \sim \ddot{z} t^2 \sim v_0^2 \frac{l^{3/2}}{r^{5/2}} t^2 \,. \tag{47}$$

In the first estimation we assumed that the elastic constants of the substrate and particle are of the same order of magnitude. This hypothetical displacement $z >> l$ as long as

$$v_0^2 \frac{l^{3/2}}{r^{5/2}} t^2 >> l \Rightarrow \left(\frac{v_0 t}{r} \right)^2 \sqrt{\frac{l}{r}} >> 1 \,. \tag{48}$$

For the typical numbers employed in ns laser cleaning (l~5 nm, r~1 μm) l.h.s. is about 10^3, i.e., condition (48) is practically always fulfilled in *ns* laser cleaning, especially with smaller particles. This means that the substrate is not appreciably slowed down by the particle. In other words, we can treat the influence of the particle on substrate expansion on the basis of the quasi-static Hertz problem. This *does not* mean that the substrate is not

deformed. The total deformation in Eq. (15) contains particle and substrate parts $h = h_p + h_s$ (see Fig. 1), which are in relation (Landau, 1986a)

$$h_p : h_s = \frac{1-s_p^2}{Y_p} : \frac{1-s_s^2}{Y_s} . \qquad (49)$$

Thus, "soft" substrates will have an indentation of the order of h, determined by Eq. (15), but not of the order of l. Note, that though we used a ***geometrical*** approximation for the contact radius a, Fig. 1 and equation (15), are based on a more realistic Hertzian picture. Within the geometrical approximation $h_p : h_s = r_s : r_p$ and the plane substrate with r_s ® ¥ is not deformed.

In *ps* cleaning one has to include the influence of the particle in the boundary conditions (31) for the substrate and the elastic problem becomes dynamic and essentially 3D even for wide beams.

3.6. Unilateral dynamic expansion

We give here for reference purposes the solution for ***dynamic*** expansion with ***free*** boundary in 1D case with constant parameters. The applicability of this solution to laser cleaning requires additional discussion, as the boundary not always can be considered as free. The derivation and consideration of sound related effects will be presented elsewhere. We introduce the longitudinal sound velocity

$$v_0^2 = \frac{Y(1-s)}{r(1+s)(1-2s)} \qquad (50)$$

and in the same notations as before rewrite Eqs. (29), (31), (32) for the 1D case in the form:

$$\ddot{u} = v_0^2(u_{zz} - b_1 T_z) \ , \ u_z\big|_{z=0} = b_1 T \ , \ u, u_z\big|_{z\to\infty} = 0 . \qquad (51)$$

Displacement l defined as in Eq. (42) is (see also (Maznev, 1997))

$$l(t) \equiv -u(0,t) = b_1 v_0 \int_0^t T(v_0(t-t_1), t_1)dt_1 = b_1 \int_0^{v_0 t} T(z, t - z/v_0)dz . \qquad (52)$$

This is an obvious generalization of static expression (38). Indeed, with $v_0 \to \infty$ we recover static result.

3.7. Thermal expansion of absorbing particle

The temperature of a ***small*** particle with thermal diffusivity D_p is homogeneous if $D_p t >> r^2$, i.e., with $r \sim 10^{-5}$ cm and $D_p \sim 0.1$ cm^2/s, for $t >>$ 1 ns. If volumetric thermal expansion coefficient of the particle is b_p, the increase in volume V is given by:

$$\dot{V} = V b_p \dot{T}_p \Rightarrow \dot{h} = \frac{V}{S} b_p \dot{T}_p = \frac{b_p}{3} r \dot{T}_p . \tag{53}$$

Here, S is the surface area of the particle and the last equality assumes spherical shape. Temperature evolution of an ***absorbing*** particle with (quasi-static) heat contact with substrate can be approximated by

$$cm\dot{T}_p = s_a I - 4K_s a(T_p - T_s) . \tag{54}$$

where $s_a \approx pr^2 A_p < pr^2$ is the total absorption cross section (for small particles diffraction effects should be considered and the expression for s_a is quite different (Born, 1980)). Here, a is contact radius, and we used the formula (8.2.10) from (Carslaw, 1959) for the flux into the semi-infinite substrate from the uniformly heated disk. Indexes p and s refer to particle and substrate respectively, T_s being the temperature of the substrate "far away from the particle". Combining Eqs. (53) and (54) we obtain for the extreme case with no heat contact

$$\dot{h} = \frac{b_p r s_a}{3 c_p m} I \approx \frac{b_p}{4 c_p r_p} I_a < \frac{b_p}{4 c_p r_p} I , \tag{55}$$

which shows similarity with Eq. (42) for the substrate expansion. In the limiting case with poor thermal contact the material with the biggest thermal expansion coefficient provides the biggest contribution to thermally induced deformation and elastic forces. The case with the substrate/particle thermal contact deserves further consideration and will be presented elsewhere.

3.8. Transparent particle heated by the substrate

Another extreme case is the transparent particle, which does not disturb the absorption of light. Or, more accurately, it does not alter the temperature field. This requirement is less restrictive for small particles, as heat conduction smoothes small-scale intensity inhomogeneities during ns pulses. Let us assume that the particle/substrate contact is so good that their temperatures are equal. This will give us an upper limit for the particle temperature. Energetic estimation (7.5.8b) from (Bäuerle, 2000) yields for the surface temperature (constant parameters):

$$f_a \approx c_s r_s T_s (l_a + l_T) \Rightarrow I_a \approx c_s r_s \Delta T_s (l_a + l_T) + c_s r_s T_s \Delta l_T \Rightarrow$$
$$\Delta T_s \approx \frac{I_a}{c_s r_s (l_a + l_T)} - \frac{f_a \Delta l_T}{c_s r_s (l_a + l_T)^2}. \qquad (56)$$

Here l_a and l_T are absorption and thermal lengths. Substituting this into Eq. (53) we obtain

$$\Delta r = \frac{b_p r}{3 c_s r_s (l_a + l_T)} \left(I_a - \frac{f_a \Delta l_T}{(l_a + l_T)} \right) \le \left(\frac{r}{l_a + l_T} \right) \frac{b_p I_a}{3 c_s r_s}. \qquad (57)$$

This expression has a similar structure as Eq. (42). The ratio $r / (l_a + l_T)$ is typically (much) less than one. Thus, particle expansion can be taken into account by a replacement $b_s \to b_s + C_1 b_p$ in Eq. (42), which modifies coefficient C in Eq. (74).

3.9. Maximum velocity of ejected particles

Let us now estimate the ***maximum*** particle velocity v if the cleaning is based on thermal expansion and elasticity. Similar estimations, but not expressed in terms of laser fluence, were done in (Lu, 2000a). We neglect the initial adhesion energy, assume that the particle does not move during the pulse and that all elastic energy is later transformed into kinetic energy (big particles or short pulses). Then the energy balance yields:

$$\frac{2\bar{Y}}{5} r^{1/2} l^{5/2} = m\frac{\dot{x}^2}{2} \Rightarrow \dot{x} \sim \left(\frac{\bar{Y}\, l^{5/2}}{r\, r^{5/2}} \right)^{1/2} \sim v_0 \left(\frac{l}{r} \right)^{5/4} . \qquad (58)$$

Here we assumed comparable material properties of particle and substrate. Including possible expansion of the particle *Dr* and using Eqs. (42), (55) and (74), we obtain with values from the Table 1 and $f_a \approx 1$ J/cm^2 an upper estimation:

$$\dot{x} \sim v_0 \left(\frac{l + Dr}{r} \right)^{5/4} \sim v_0 \left(C \frac{b f_a}{c r r} \right)^{5/4} \sim 10^4 \text{ cm/s} . \qquad (59)$$

As in most cases l, $Dr << r$, the velocity of the ejected particles is always rather small. It is even smaller for small particles, as they move as a whole during the expansion. Elastic mechanisms yield higher velocities. Indeed, the velocity of ***non-deformable*** particles cannot exceed that of the moving surface given by Eq. (42) or, better by Eq. (74):

$$\dot{x} \sim C \frac{b I_a}{c r} \sim 0.25 \times 10^3 \text{ cm/s} . \qquad (60)$$

Results for intermediate particle sizes can be obtained by solving Eq. (15) as explained in sections 4,5. If measured particle velocities (Schrems, 2000) significantly exceed both Eqs. (59) and (60), this is a strong indication that other mechanisms are responsible for cleaning.

4. Cleaning threshold

4.1. General threshold conditions

Let us formulate the cleaning condition for the general law of particle movement:

$$\ddot{h} + g\dot{h} + \frac{1}{m}\frac{\partial U}{\partial h} = \dot{v} . \qquad (61)$$

Here $\dot{v}$ in the r.h.s. may include expansion of both substrate and particle (see Eq. (15)) or other cleaning forces, while U may include capillary effects, etc. We neglect initial and escape velocities, and define

$U_{ad} = U(t=\infty) - U_0$ (difference between escape and initial energies). Multiplying Eq. (61) by $\dot{h}$ and integrating we obtain the energy criterion

$$\int_0^\infty \dot{v}\dot{h}dt' > m^{-1}U_{ad} + g\int_0^\infty \dot{h}^2 dt' \ . \tag{62}$$

The l.h.s. is the (specific) work of the cleaning force, while the second term on the r.h.s. is the dissipative loss caused by damping. To write the threshold condition in terms of fluence, one has to solve equation of motion (61). Some limiting cases allow more general consideration.

4.1.1. Short cleaning pulse

If the pulse is short i.e., $t << t_0$, g^{-1} cleaning force $m\dot{v}$ dominates *during the action of the pulse*. Neglecting damping and potential in Eq. (61) we obtain:

$$\dot{h} \approx v \Rightarrow h(t) \approx h_0 + \int_0^t v\,dt, \ \ \dot{h}(t) \approx 0\,. \tag{63}$$

Thus, energy acquired at the end of the pulse is due to change in h (deformation). If damping is week ($g << w_0$), cleaning will take place (after the pulse) if the accumulated (potential) energy is higher than the detachment energy. This is ***elastic energy*** cleaning regime. For the potential (5) with expansion of both substrate and particle taken into account, this results in

$$\begin{aligned} &U(h_0 + \int_0^t v\,dt) > 0, \ \ \int_0^t v\,dt = l + Dr \Rightarrow \\ &l + Dr > \left((5/2)^{2/3} - 1\right)h_0 \approx 0.84h_0 \sim h_0. \end{aligned} \tag{64}$$

Last approximation refers to a parabolic potential (9).

4.1.2. Long cleaning pulse

If the pulse is long, i.e., $t >> t_0$, one can solve Eq. (61) in a quasi-static approximation. Internal oscillations are weakly excited because there are no high harmonics in the spectrum of the cleaning force. As a result h is

determined by the condition that cleaning force $m\ddot{h}$ balances the force from the adhesion potential.

$$\frac{1}{m}\frac{\partial U}{\partial h} \approx \ddot{h}. \tag{65}$$

Thus, to clean, one has to overcome the biggest adhesion force *during the pulse*. This is *force (inertia)* cleaning regime. For the potential (5) the force is maximal with $h = 0$ and is positive in our notations. This results in:

$$(-m\ddot{h})_{\max} = -m(\ddot{l} + \ddot{w})_{\max} > F_0. \tag{66}$$

Detailed analysis shows that there exists a coefficient $C_1 < 1$ in the r.h.s., which takes into account weak internal oscillations. Its value depends on the pulse shape and g. The l.h.s. should be positive, i.e., for the mechanism based on thermal expansion, detachment occurs *in the deceleration* phase (Dobler, 1999) due to the inertia of the already accelerated particle.

4.1.3. Over-damped movement

If damping is strong ($g >> t^{-1}, w_0$), one can neglect "inertia" $\ddot{h}$ in Eq. (6). Then

$$\dot{h} \approx g^{-1}\left(\ddot{w} - \frac{1}{m}\frac{\partial U}{\partial h}\right). \tag{67}$$

Thus, as with long pulses, one has to overcome the biggest adhesion force *during the pulse*.

4.1.4. Long pulses with steep fronts

Consider a long pulse ($t >> t_0$), which starts abruptly, so that v rises to v_f within time $t_f << t_0$. Then, from Eq. (63) particle "instantaneously" acquires "velocity" $\dot{h} \approx v_f$ towards the substrate. During the rest of the pulse the position h changes weakly. If the kinetic energy associated with $\dot{h}$ exceeds that of adhesion, the particle will detach. This is the *kinetic energy* cleaning regime with the criterion:

$$mv_f^2/2 > U_{ad}\,. \tag{68}$$

Similar consideration applies for the trailing edge of the pulse. In this case the particle acquires the velocity away from the substrate. In other words, to produce strong "force" the pulse should not necessarily be short. It is enough if it has sharp edges. The criterion (68) is often less restrictive than Eq. (66).

To obtain compact analytical results for the transitional stages we consider the following problem.

4.2. Single sinusoidal pulse in parabolic potential without damping

Let us neglect damping and use a parabolic approximation for the potential. For convenience we introduce $h_1 = h - h_0$ and count the potential energy from the bottom of the well where $h_1 = 0$. The equation of motion and initial conditions become:

$$\ddot{h}_1 + w_0^2 h_1 = v\dot{}, \tag{69}$$

$$\dot{h}_1(0) = h_1(0) = 0\,. \tag{70}$$

With these notations detachment occurs when $h_1 < -h_0$.

For the sake of generality and simplicity we consider the following "sinusoidal cleaning velocity":

$$v = \frac{lw}{2p}(1-\cos wt)\,,\ 0 < t < t\,. \tag{71}$$

Here, l is the *total* displacement during the pulse, and $t \equiv 2p/w$ the *total* pulse duration with $t_{FWHM} = p/w$. Parameters l and t are convenient characteristics of the expansion process. For laser cleaning v is proportional to the laser intensity and l to the laser fluence, see Eq. (42). Thus, Eq. (71) implies similar temporal profile of the laser pulse. Qualitative results are similar for any smooth pulse. This model problem retains important features of the original formulation. At the same time it allows complete theoretical exploration and compact formulas for the main relationships between the parameters.

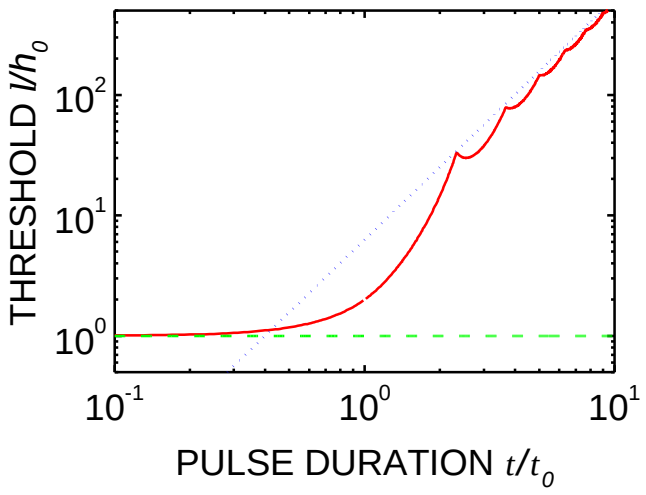

Fig. 4. Dimensionless threshold condition for a *cosinusoidal* pulse (71) and *parabolic* potential (9). Solid line - general threshold condition (73). One can see (week) resonance effects. Dashed line - short pulse limit, and dotted line - long pulse limit approximations from Eq. (73).

It is convenient to introduce the variable

$$y \equiv \frac{w_0}{w} = \frac{t}{t_0}, \tag{72}$$

where t_0 is the resonant period of the oscillator. As shown in Appendix B, the threshold condition reads:

$$\frac{l}{h_0} > \begin{cases} \dfrac{py(1-y^2)}{\sin py} & \text{for } y<1 \text{ and } \approx 1 \text{ for } y<<1, \\ -\dfrac{2py(y-1)}{\sin\left(\dfrac{2p}{y+1}\left[\dfrac{3}{4}(y+1)\right]\right)} & \text{for } y>1 \text{ and } \approx 2py^2 \text{ for } y>>1. \end{cases} \tag{73}$$

Approximate expressions in Eq. (73) are convenient for fast estimations. The result (73) is shown in Fig. 4 together with both limiting cases. With $t \sim t_0$ neither of approximations is accurate enough. One can see characteristic kinks that occur due to resonance effects. But as the cleaning force Eq. (71) contains only *one* period, these kinks are weak and can hardly be observed in experiments.

4.3. Dependence of cleaning threshold on particle radius and pulse duration

Let us rewrite threshold condition (73) in terms of the particle radius r, pulse duration t, and fluence f. For definiteness we use Eq. (6) for h_0 and Eq. (10) for w_0. The overall expansion is (see Eqs. (42) and (53))

$$l + Dr = \left(\frac{1+s_s}{1-s_s} \frac{b_s A_s}{3c_s r_s} + \frac{b_p A_p}{4c_p r_p} \right) f \approx C \frac{bA}{cr} f. \tag{74}$$

Here, the first term refers to the substrate and the second to the (absorbing) particle without thermal contact. For the transparent particle with thermal contact the second term will be of the order of Eq. (57). The last expression is a notation used for brevity. The contribution from the material with the biggest bA value dominates. C~0.25-1 is dimensionless coefficient. We neglect particle influence on the expansion of the substrate, i.e., assume that the heat conduction homogenizes the temperature near the particle and neglect field enhancement effects (Mosbacher, 2001; Leiderer, 2000; Luk'yanchuk, 2000, 2002). Rewriting the two limiting cases in Eq. (73) in dimensional quantities, we obtain the expressions for the threshold fluence:

$$f > C^{-1} \frac{cr}{bA} \begin{cases} \left(2pj/\bar{Y}\right)^{2/3} r^{1/3} & \text{short pulses or small particles} \\ & w_0 t << 1, \text{ "elastic energy",} \\ \dfrac{9t^2}{10p} \dfrac{j}{r_p r^2} & \text{long pulses or small particles} \\ & w_0 t >> 1, \text{ force (inertia).} \end{cases} \tag{75}$$

Intermediate regimes for $w_0 t \sim 1$ can be calculated numerically. The expression (73) recalculated into dimensional variables is shown in Fig. 7 by the dotted line, together with the results of more accurate calculations described in section 5.

The dependence on pulse duration is monotonic -- shorter pulses are more favorable for fixed fluence. With pulse durations shorter than the resonant period t_0, a further decrease in pulse duration is *not* advantageous. With $a\ v_0\ t \pounds 1$ one has to consider sound effects in the substrate, which will be discussed elsewhere.

The dependence on particle radius is less trivial. As w_0 and h_0 depend on r, threshold dependence on r is non-monotonic. There exists an optimal radius for a given pulse duration. For this radius the resonant period t_0 is close to the duration of the laser pulse t.

With big radii, the native period t_0 is long and the cleaning pulse is much shorter than one cycle of oscillations. Cleaning proceeds in the "elastic energy" regime. Heavy particles almost do not move during the pulse. The substrate surface moves much faster than the center of the particle. This leads to an increase in elastic energy (compression of substrate and particle). Detachment occurs after the pulse, in the first backward swing of the (internal) oscillation. This regime (for non-linear potential) is shown in Fig. 5 a. For a symmetric parabolic potential the elastic energy become positives in the compression stage if $l > h_0$, and this yields simplified detachment condition for such a potential. A more accurate estimation (64) for non-linear potential does not change the threshold significantly. This can be seen also from the comparison of the solid and dotted curves in Fig. 7. The $r^{1/3}$ increase in threshold with radius is due to bigger equilibrium value of h_0 and higher adhesion energy $|U_0|$ in Eq. (7) for bigger particles.

With smaller particles, the native period becomes shorter than the laser pulse duration. The response of the oscillator to the "low frequency" force is inefficient. Cleaning proceeds in the "quasi-static" regime, when fast and small internal oscillations in h are superimposed on the slow changes in h that obey Eq. (65). This regime is shown in Fig. 5b. Let us derive the threshold condition for this situation from the general expression (66). For the pulse (71) the largest deceleration occurs at $3t/4$, and in notations (74) the cleaning condition is:

$$mC\frac{bA}{cr}(-\ddot{h})_{\max} \equiv \left(\frac{4\pi r^3}{3}\rho_p\right)C\frac{bAf}{cr}\frac{2\pi}{t^2} > F_0 \equiv 2\pi r\sigma \Rightarrow$$

$$f > C^{-1}\frac{cr}{bA}\left(\frac{3t^2}{4\pi}\frac{\sigma}{\rho_p r^2}\right) \tag{76}$$

The results (75) and (76) slightly differ because the pull-out force $F_0=(6/5)2\pi r\sigma$ in parabolic approximation. The strong increase in the threshold fluence with smaller particles demonstrates the inefficiency of the "force (inertia)" cleaning regime.

Let us consider kinetic energy cleaning regime (68). The movement of the particle for a rectangular laser pulse is shown in Fig. 5c. If intensity change at the steep front is I_f,

$$\frac{m}{2}(\dot{h}+\dot{l})^2 \equiv \frac{4\pi r^3}{6}\rho_p\left(C\frac{bAI_f}{cr}\right)^2 > |U_0| \equiv \frac{3}{5}(2\pi\sigma)^{5/3}\bar{Y}^{-2/3}r^{4/3} \Rightarrow$$
$$I_f > C^{-1}\frac{cr}{bA}\left(\frac{3(2\pi)^{1/3}}{5^{1/2}}\frac{\sigma^{5/6}}{\rho_p^{1/2}\bar{Y}^{1/3}r^{5/6}}\right). \tag{77}$$

This can be ***formally*** written as a condition for fluence. For example assuming $I_f \approx f/t$ we get

$$f > C^{-1}\frac{cr}{bA}\left(\frac{3(2\pi)^{1/3}}{5^{1/2}}\frac{\sigma^{5/6}t}{\rho_p^{1/2}\bar{Y}^{1/3}r^{5/6}}\right) \tag{78}$$

But with steep fronts it is the ***intensity*** what is important. Note also much weaker dependence of threshold on r as compared with Eq. (76). This is crucial for small particles.

With $t \sim t_0$ no simple approximations for the threshold exists. This situation is shown in Fig. 6. Together with h and l, the movement of the particle center in the laboratory frame $z - z_0$ is shown. In the beginning, surface displacement l is faster than the particle movement (compression) and later the particle detaches with constant velocity.

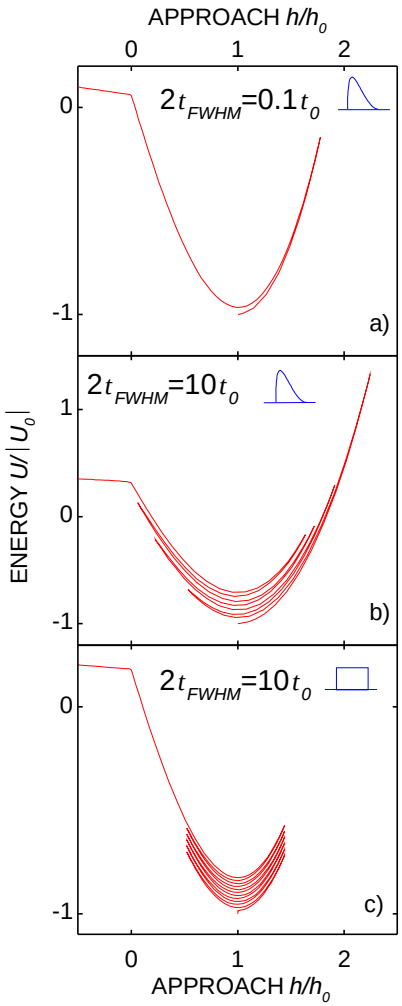

Fig. 5. Movement of the particle (evolution of the approach h in non-linear potential (5)) under the effect of substrate expansion somewhat above threshold. Temporal profile of the laser pulse is given by Eq. (44). a) Elastic energy cleaning regime for big particle. Laser pulse is much shorter than the period of oscillator $2t_{FWHM} = 0.1t_0$ b) Quasi-static force/inertia regime for small particle. Laser pulse is much longer than the period of oscillator $2t_{FWHM}=10t_0$. c) Kinetic energy regime for the pulse with steep fronts. Rectangular laser pulse is longer than the period of oscillator $2t_{FWHM} = 10\ t_0$, while rise/fall time of the fronts $t_f << t_0$.

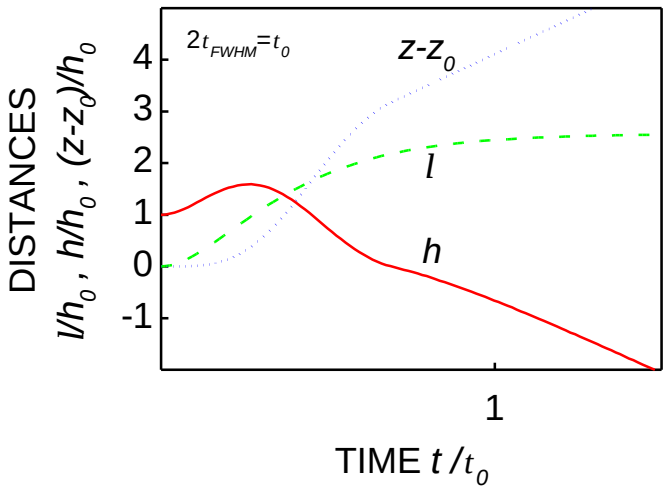

Fig. 6. Movement of the particle under the effect of substrate expansion. Potential and pulse shape as in Fig. 5. Parameters are such, that $2t_{FWHM} = t_0$ and cleaning threshold is slightly exceeded. Solid line - evolution of the approach h. Dashed line - surface expansion l. Dotted line - movement of the particle center z in the laboratory reference frame (defined in Eq. (14)) referred to its initial position z_0.

5. SiO_2 particles cleaned from Si wafers

Let us now study laser cleaning of SiO_2 particles from Si surfaces. The goal is to compare the predictions of the theory with experimental investigations. In order to diminish the influence of humidity and to avoid redeposition the experiments were performed in vacuum.

5.1. Experimental

The cleaning of SiO_2 particles (Bangs Laboratories, radii 200-2585 nm) from (100) Si wafers (Wacker Siltronic) was performed with a KrF excimer laser (Lambda Physik LPX 205, wavelength 248 nm, pulse duration 31 ns FWHM). The energy of the beam was controlled by an external attenuator (tilted quartz plate) and projected with a mask onto the target to a spot of 1 mm diameter. Such an imaging produces a uniform energy distribution within the irradiated area. The pressure within the chamber was ~ 4×10^{-5}

mbar. Particles are deposited onto the sample by spin-coating, which gives a high uniformity of the particle density. Optical microscopy and picture processing software, which can count the particles and measure their size, is used to evaluate the cleaning efficiency. Since the homogeneity of the samples is not perfect, a picture of the cleaned area is taken before irradiation and is compared with the picture after irradiation. With this technique it is possible to see the behaviour of clusters and redeposition of the particles.

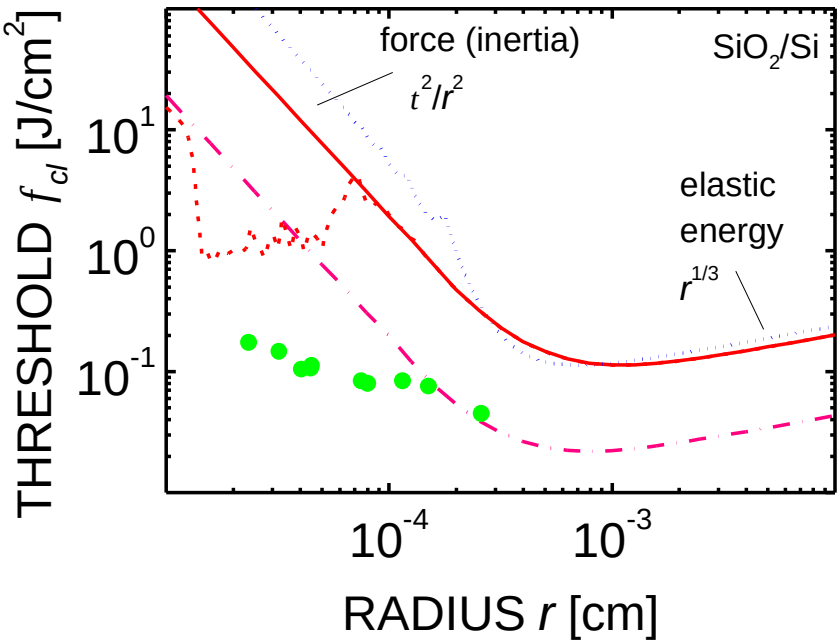

Fig. 7. Experimental and calculated cleaning fluence as a function of particle radius for SiO_2 particles on Si. Parameters used in the calculations are listed in Table 2. Only expansion of the substrate is taken into account. Circles – experimental points. Solid line - numerically calculated threshold. Dash-dotted line - numerically calculated threshold for ten times smaller adhesion. Resonance effects for the considered *anharmonic* potential (5) and *realistic* pulse shape (44) are absent. Dotted line - recalculated into dimensional quantities harmonic approximation (73) for the same t_{FWHM}. Dashed line - numerically calculated threshold for "rigged" laser pulse (80).

5.2. Cleaning threshold vs. radius

Theoretical and experimental results are compared in Fig. 7. The parameters used in the calculations are listed in Table 2. Their choice is somewhat arbitrary. For example Si is rather anisotropic, vitreous SiO_2 does not accurately follow the Grüneisen relation, etc. Because it is more difficult to take into account thermal expansion of the particle (see section 3), only expansion of the substrate is included. This is justified, as expansion of ***fused*** silica is much smaller than that of silicon (Table 2) and should not significantly alter the results. Damping was ignored.

Table 2. Parameters used in the calculations. For strongly temperature-dependent parameters values for room and highest available temperature are given with temperature (in K) indicated in brackets. For weakly varying parameters value at or somewhat above room temperature is taken. Some of the elastic properties vary in the literature and for crystal may depend on direction. Average values are taken in this case.

Parameter	**Value(s)**	**Ref.**	**Comments**
Pulse duration t ns	12.7		31 t_{FWHM}
Laser wavelength l nm	248		KrF laser
Substrate Si			
Specific heat c_s J/gK	0.72 (300) 1 (1500)	(Bäuerle, 2000)	used value
Volumetric thermal expansion coefficient b_s K^{-1}	7.7×10^{-6} (300) 13.2×10^{-6} (1400)	(Landolt, 1982)	used value
Poisson ratio s_s	0.26-0.28 0.27	(Flina; Almaz; Landolt, 1982)	anisotropic used value
Young modulus Y_s dynes/cm^2	$1.3\text{-}1.9\times10^{12}$(300) $1.2\text{-}1.8\times10^{12}$(900) 1.6×10^{12}	(Landolt, 1982)	100-111 direction used value v_s=9.13×10^5 cm/s (110)
Density r_s g/cm^3	2.3	(Bäuerle, 2000)	used value
Absorption coefficient a_s cm^{-1}	1.67×10^6	(Bäuerle, 2000)	*weak a(T)*
Absorptivity A_s	0.39	(Bäuerle, 2000)	*weak A(T)*
Melting temperature T_{ms} K	1690	(Bäuerle, 2000)	

Table 2. (Continuation)

Parameter	**Value(s)**	**Ref.**	**Comments**
Particle SiO_2 (fused silica)			
Specific heat c_p J/gK	0.72 (300) 1.22 (1000) 1	(Bäuerle, 2000)	 used value
Volumetric thermal expansion coefficient b_p K^{-1}	1.65×10^{-6} (300) 1.2×10^{-6} (300) 1.8×10^{-6} (500) 1.4×10^{-6} (1100) 1.65×10^{-6}	(Prokhorov, 1990) (Gray, 1972)	(300-1300) used value
Poisson ratio s_p	0.17	(Goodfellow; Weast, 1989)	used value
Young modulus Y_p dynes/cm^2	$0.7\text{-}0.75\times10^{12}$ 0.73×10^{12}	(Goodfellow; Weast, 1989)	 used value $v_s = 5.9\times10^5$ cm/s
Density r_p g/cm^3	2.2	(Bäuerle, 2000)	probably smaller (Bangslabs)
Absorption coefficient a_p cm^{-1}	1	(Bäuerle, 2000)	
Absorptivity A_p	0.94	(Bäuerle, 2000)	
Melting temperature T_{mp} K	1873	(Bäuerle, 2000)	
Adhésion			
LVdW constant $V_{Si\text{-}Si}$*=(4p/3)H, eV* *H* - Hamaker constant	6.5-6.76 6.8-7.2 6.15 6.7	(Visser, 1976) (Bowling, 1989) (Dahneke, 1972)	 used value
LVdW constant $V_{SiO2\text{-}SiO2}$*=(4p/3)H, eV* *H* - Hamaker constant	1.7 1.9-12 2.33 1.32 2	(Bergström, 1997) (Visser, 1976) (Dahneke, 1972) (Heim, 1999)	 assuming DMT (Derjaguin, 1975) used value
LVdW constant *eV* $V_{Si\text{-}SiO2} = (V_{Si-Si}\ V_{SiO2\text{-}SiO2})^{1/2}$	3.66		used value
Adhesion distance *e* cm	1.41×10^{-8}	(Visser, 1976; Bowling, 1989)	rather universal
Work of adhesion j erg/cm^2	140		used value

We used average parameters for Si and fused SiO_2 available in the literature and calculated the adhesion between them according to the formula (Visser, 1976)

$$j_{12} = \sqrt{j_{11} j_{22}} \quad . \tag{79}$$

But one has to have in mind, that

- In the presence of the native oxide the contact is essentially SiO_2-SiO_2, which reduces j by about a factor of two.
- Several monolayers of moisture trapped in the particle-substrate interstice during the spin-on procedure can decrease adhesion by an order of magnitude due to screening of VdW interaction by water, which has a high dielectric constant.
- We *do not* discuss here influence of capillary effects on adhesion. One has to have in mind, however, that with elevated temperatures induced in laser cleaning, surface tension coefficient significantly decreases, making capillary forces less important.
- Likewise not discussed here is increase in adhesion due to possible charge of particles. This can be included into consideration, but is usually more important for bigger particles and/or in the post-detachment stage.
- Adhesion may increase with the storage time due to formation of covalent bonds and possible plastic deformation even for rather hard materials.
- Surface roughness of substrate *and/or* particle can further change adhesion.

Having this in mind, we present calculations also for the adhesion decreased by a factor of 10. The result, and dependence on other parameters, can be understood from expressions (75). Though it is more difficult to clean smaller particles, experimentally observed thresholds (circles) and the slope of $f_{cl}(r)$ dependence are much *lower* than theory predicts. Even small adhesion cannot explain these findings. What can be the reason for such a behavior?

5.3. Role of small oscillations in intensity

One possibility is the "bad quality" of the excimer laser pulse. Typical excimer pulses are too long so that the small particles are removed in the

inefficient "force" regime. If the pulse contains high frequency components, they may significantly reduce the threshold for small particles, despite small fluence contained in each "spike". Numerically calculated threshold for the rigged pulse of the same total fluence is shown by the dashed curve. The rigged pulse had a temporal profile with harmonics characterized by j.

$$I(t) = \frac{2}{\{(j_{\max} - j_{\max})/dj\} + 1} \left(\sum_{j=j_{\min},\, dj}^{j_{\max}} \cos^2\left(2pj\frac{t}{t} + j^3 \right) \right) I_0 \frac{t}{t} \exp\left(-\frac{t}{t} \right). \quad (80)$$

Here the integer part {} in the denominator makes the overall fluence independent on j_{min} , j_{max} and dj, which were taken as j_{min} = 1.1415, j_{max} = 5, and dj = 0.2718. The surface displacement, velocity, and acceleration are shown in Fig. 8. The displacement is virtually the same as for the smooth pulse (Fig.3). Velocity (which is proportional to intensity) and especially acceleration differ significantly. Short spikes in the intensity may resonantly decrease the threshold for small particles by two orders of magnitude.

Another reason, which seem to be more plausible (Leiderer, 2000; Lu, 2000b) may be field enhancement by the particles (Mosbacher, 2001; Luk'yanchuk, 2000, 2002; Zheng, 2001; Lu, 2000c) or explosive vaporization of residual moisture (Fourrier, 2001).

5.4. Suggestions for cleaning experiments

Oscillations of adhering particles may be used to increase efficiency of DLC. One can try to utilize possible resonance effects with the aim to remove smaller particles and to increase damage-free cleaning window. We discuss several possibilities.

Smooth excimer laser ns pulse is "too long" for sub-μm particles. If it is modulated with the frequency that matches internal "adhesion frequency" (10) for the particles of given size, one can expect resonance increase in oscillation amplitude. Calculations demonstrate that if the overall duration of the pulse stays constant, and the period of the oscillations is about one tenth of the overall pulse duration, cleaning threshold can decrease by 1-2 orders of magnitude. Due to the non-linearity of the potential (5), even without damping, at near threshold fluences resonance growth "saturates" after 5-10 oscillations. Detailed investigation of this effect will be presented

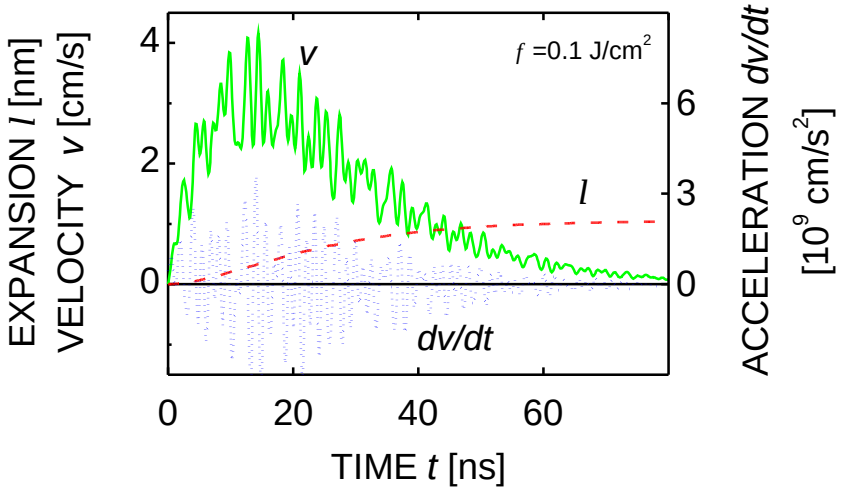

Fig. 8. Surface displacement *l* (dashed line), velocity *v* (solid line) and acceleration *dv/dt* (dotted line) for the temporal profile of rigged pulse (80) used for the calculations of dashed line in Fig. 7. All other parameters are as in Fig. 3. Note difference in scale for acceleration as compared to Fig. 3.

elsewhere (Arnold, 2002 a, b).

Without damping the cleaning effect of the pulse of duration $t = t_0$ (***single resonant*** "push") and longer ***modulated*** pulse which includes n ***resonant*** "pushes" $t = n\ t_0$ is similar if they have ***the same overall fluence***. But heating will be lower for the longer pulse, proportionally to $n^{-1/2}$ for surface absorption (Bäuerle, 2000). Thus, damage threshold will increase and the window for damage-free cleaning may widen.

"Infinitely short" pulse is the most efficient for the given fluence. It is more efficient than the modulated pulse of arbitrary duration. But with short pulses damage threshold is determined by l_a and is much lower than for ns pulses (Bäuerle, 2000). One can replace one short pulse with several pulses with the fixed delay between them. Mode locked lasers are natural candidates for such experiments. If the delay matches internal frequency of the oscillations, the ***cleaning*** effect will be the same. ***Damage*** threshold will be determined rather by the ***overall*** duration t of the pulse train, provided that $l_T \sim (D_s t)^{1/2} >> l_a$. Note, that the description of ps laser cleaning

requires consideration of sound related effects that become important with $a\ v_0\ t < 1$ or $v_0 t\ /r < 1$.

6. Conclusions

In this article we have provided a theoretical analysis of dry laser cleaning using ns pulses. Expressions for the thermal expansion of the substrate are derived and discussed for different situations. The formula for the 1D quasi-static thermal expansion of the substrate does not require solution of the heat equation and is valid over a broad range of temperature-dependent material parameters. The expansion of absorbing and transparent particles is discussed as well.

A simple approximation for a combined elastic-VdW potential has been suggested. The laser cleaning process is formulated as an escape problem from the non-linear potential under the action of cleaning force produced by thermal expansion. Expansion of the substrate and the particle are treated on a unified basis. Possible damping mechanisms are discussed.

Two parameters characterize the adhesion potential -- the period of oscillations near the bottom of the potential well t_0 and the equilibrium deformation (approach) h_0. They serve as natural temporal and spatial scales. Their analytical dependence on particle size r and material properties is provided. Laser pulse duration t should be compared with t_0 and overall thermal expansion $l+Dr$ with h_0.

Formulas for the cleaning fluence f_{cl} in different regimes are derived and compared with numerical calculations. In particular, with $t < t_0$ (large particles) cleaning proceeds in the "elastic energy regime" which reduces to the condition $l+Dr > h_0$. As a result, $f_{cl} \propto r^{1/3}$. With $t > t_0$ (small particles) cleaning proceeds in the inefficient "force regime" and $f_{cl} \propto t^2/r^2$, which favors shorter laser pulses. With $t > t_0$, but steep edges of the pulse $t_f << t_0$, cleaning requires that particle kinetic energy exceeds that of adhesion $m(\dot{l} + D\dot{r})_f^2 / 2 > |U_0|$. This leads to $f_{cl} \propto t\ /r^{5/6}$ for the "kinetic energy" regime.

Comparison with experimental $f_{cl}(r)$ dependence for SiO_2 particles on Si surface shows that commonly assumed mechanisms of dry laser cleaning do not explain experimental findings. Experimentally observed thresholds are *too low*. Among possible explanations are fast spatial-temporal

variations in intensity of excimer (KrF) laser pulse and field enhancement effects suggested earlier by other authors.

Utilization of resonance effects either by modulation of ns laser pulse or employing the train of ps pulses with delay equal to $t_0(r)$ is suggested. Developed approach can be applied to the cases when other adhesion forces (capillary, electrostatic, chemical bonding, etc.) may dominate.

Acknowledgments

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Appendix A. Quasi-static 3D thermal expansion

Here we derive the surface displacement for the semi-infinite substrate $z > 0$. The idea is to write equations and boundary conditions for u_z and $div\ \boldsymbol{u}$ only (more accurately for some function f introduced below), and to solve them by Fourier transform in x-y plane. Henceforth ∂_z denotes derivative with respect to z, while index z refers to the component of a vector. Stationary equation (29) for z component can be written as:

$$Du_z + \frac{1}{1-2s}\partial_z div\,\boldsymbol{u} - \frac{b}{3}\frac{2(1+s)}{1-2s}\partial_z T = 0\,. \qquad \text{(A.1)}$$

At the same time, applying div to the stationary equation (29) we get

$$D(div\,\boldsymbol{u} - b_1 T) = 0\,. \qquad \text{(A.2)}$$

Boundary conditions (31) also can be written in terms of ***div u*** and u_z only. They are valid in ***x-y*** plane $z = 0$ and can be differentiated in this plane. The following combination does not contain u_x and u_y separately, and can be used as a boundary condition for u_z

$$\partial_x s_{xz} + \partial_y s_{yz} = \frac{Y}{2(1+s)}\left(\partial_{xx} u_z + \partial_{yy} u_z - \partial_{zz} u_z + \partial_z div\,\boldsymbol{u}\right) = 0 \text{ at } z = 0 . \quad \text{(A.3)}$$

To deal only with the first order boundary conditions, we exclude $\partial_{zz} u_z$ using Eq. (A.1).

$$D_\perp u_z + \frac{1-s}{1-2s}\partial_z div\,\boldsymbol{u} - \frac{b}{3}\frac{1+s}{1-2s}\partial_z T = 0 \text{ at } z = 0 . \quad \text{(A.4)}$$

Here $D_\perp$ stands for 2D Laplacian in ***x-y*** plane. Second boundary condition is Eq. (31) for normal stress s_{zz} with definition Eq. (32).

$$s_{zz} = 0 \Rightarrow \partial_z u_z + \frac{s}{1-2s} div\,\boldsymbol{u} - \frac{b}{3}\frac{1+s}{1-2s} T = 0 \; at \; z = 0 . \quad \text{(A.5)}$$

The variable more convenient than ***div u*** is ***f*** defined as

$$f = div\,\boldsymbol{u} - b_1 T . \quad \text{(A.6)}$$

Rewriting equations (A.2) and (A.1) in terms of ***f*** we get:

$$\Delta f = 0, \qquad \Delta u_z + \frac{\partial_z f}{1-2s} - b_1 \partial_z T = 0. \quad \text{(A.7)}$$

And for the boundary conditions (A.4) and (A.5) at $z = 0$ and at infinity we obtain

$$\Delta_\perp u_z + \frac{1-s}{1-2s}\partial_z f = 0 \quad \text{at } z = 0,$$

$$\partial_z u_z + \frac{s}{1-2s} f - b_1 T = 0 \text{ at } z = 0, \tag{A.8}$$

$$u_z,\ f,\ T \to 0 \qquad \text{at } z \to \infty.$$

These coupled equations are solved by Fourier transform in *x-y* plane. There exist subtle requirements, that all quantities (including *f*, which is a difference of two "good" functions, see Eq. (A.6)) disappear at infinity and can be Fourier transformed. The former property holds in physically admissible situations. The latter is more restrictive. It is *not* satisfied for the stationary temperature distribution in the semi-infinite substrate induced by a permanent finite source. In this case *T~1/r* at large distances and Fourier transform *of displacement* u_z does not exist. This is the mathematical reason why one cannot obtain unilateral expansion (42) as a limiting case of the formulas from this appendix. With time dependent temperature distributions induced by spatially finite sources present results should be used.

The Fourier transform of Eq. (A.7) (taking into account conditions at infinity) with wave vector $\boldsymbol{k}$ (length k), results in trivial equation for *f*, which can be immediately solved, and in differential equation for u_z

$$\begin{gathered}\tilde{f} = \tilde{f}_0 e^{-kz}, \\ \partial_{zz}\tilde{u}_z - k^2\tilde{u}_z - \frac{k\tilde{f}_0 e^{-kz}}{1-2s} - b_1\partial_z\tilde{T} = 0.\end{gathered} \tag{A.9}$$

Here, tilde denotes Fourier image and f_0 º f (z=0). Transformed boundary conditions (A.8) look like

$$\begin{aligned}&k^2\tilde{u}_z + \frac{1-s}{1-2s}k\tilde{f}_0 = 0 \qquad \text{at } z = 0, \\ &\partial_z\tilde{u}_z + \frac{s}{1-2s}\tilde{f}_0 - b_1\tilde{T} = 0 \text{ at } z = 0, \\ &\tilde{u}_z \to 0 \qquad \text{at } z \to \infty.\end{aligned} \tag{A.10}$$

We find $\tilde{f}_0$ from the first equation and exclude it from the remaining boundary condition *and the equation* for u_z. Introducing $\tilde{u}_z(0) \equiv \tilde{u}_z(z=0)$ we get the equation for the Fourier image of u_z only.

$$\begin{gathered} \partial_{zz}\tilde{u}_z - k^2\tilde{u}_z + \frac{k^2\tilde{u}_z(0)}{1-s}e^{-kz} - b_1\partial_z\tilde{T} = 0, \\ \partial_z\tilde{u}_z(0) - \frac{sk\tilde{u}_z(0)}{1-s} - b_1\tilde{T}(0) = 0\ , \quad \tilde{u}_z(\infty) \to 0. \end{gathered} \tag{A.11}$$

This linear equation can be solved in the general case. Expression that satisfies condition at infinity is:

$$\begin{aligned} \tilde{u}_z = \frac{1+2kz}{4(1-s)}\tilde{u}_z(0)e^{-kz} + \frac{b_1}{2k}e^{kz}\int_{\infty}^{z} e^{-kz_1}\tilde{T}(z_1)dz_1 - \\ - \frac{b_1}{2k}e^{-kz}\int_{0}^{z} e^{kz_1}\tilde{T}(z_1)dz_1 + c_1e^{-kz}. \end{aligned} \tag{A.12}$$

Coefficient c_1 is from the solution of homogeneous equation (decaying at $z \to \infty$). This solution should be self-consistent, i.e., $\tilde{u}_z(z=0) = \tilde{u}_z(0)$, and it should satisfy boundary condition in Eq. (A.11). This results in:

$$\begin{gathered} \frac{1}{4(1-s)}\tilde{u}_z(0) + \frac{b_1}{2k}\int_{\infty}^{0} e^{-kz_1}\tilde{T}(z_1)dz_1 + c_1 = \tilde{u}_z(0), \\ k\left(-c_1 + \frac{1-4s}{4(1-s)}\tilde{u}_z(0)\right) + \frac{b_1}{2}\left(\int_{\infty}^{0} e^{-kz_1}\partial_z\tilde{T}(z_1)dz_1 - 2\tilde{T}(0)\right) = 0. \end{gathered} \tag{A.13}$$

Resolving this couple of equations for c_1 and $\tilde{u}_z(0)$, substituting b_1 from Eq. (34) and performing integration by parts we find:

$$\tilde{u}_z(0) = \frac{2b(1+s)}{3k}\left(\int_{\infty}^{0} e^{-kz_1}\partial_z\tilde{T}(z_1)dz_1 - \tilde{T}(0)\right) = \frac{2b(1+s)}{3}\int_{\infty}^{0} e^{-kz_1}\tilde{T}(z_1)dz_1\ . \tag{A.14}$$

$$c_1 = \frac{b(1+s)}{3k(1-s)}\left((1-2s)\int_{\infty}^{0} e^{-kz_1}\partial_z \tilde{T}(z_1)dz_1 - \frac{3-4s}{2}\tilde{T}(0)\right) =$$
$$= b_1\left((1-2s)\int_{\infty}^{0} e^{-kz_1}\tilde{T}(z_1)dz_1 - \frac{\tilde{T}(0)}{2k}\right).$$

Let us give for reference purposes the resulting compact expression for $\tilde{u}_z$

$$\tilde{u}_z = b_1\left[\left(\frac{3-4s}{2}+kz\right)e^{-kz}\int_{\infty}^{0} e^{-kz_1}\tilde{T}(z_1)dz_1 + \right.$$
$$\left. + \frac{1}{2}\left(e^{-kz}\int_{0}^{z} e^{kz_1}\tilde{T}(z_1)dz_1 + e^{kz}\int_{\infty}^{z} e^{-kz_1}\tilde{T}(z_1)dz_1\right)\right]. \qquad \text{(A.15)}$$

These expressions can be useful for dry laser cleaning problem with tightly focused beams, or with local field enhancement under the particle. In both cases the source term in the heat equation and temperature distribution are 3D, but ***elasticity*** can be considered quasi-statically. The approach developed here may prove useful also for the time-dependent elasticity. Similar considerations are known in acoustic studies (Dubois, 1994) with more complicated problems, but there numerical calculations were heavily involved. Our presentation provides compact, closed form results, especially for the measurable surface displacement.

One can simplify the results for given temperature distribution, or write the equation for the Fourier image of temperature from the heat equation and relate the displacement directly to the source term (or its Fourier image). This will be considered elsewhere. General result for ***non-transformed*** quantities can be obtained if temperature distribution is almost 1D, but is nevertheless limited in ***x-y*** directions. In this case stresses, strains, ***and displacements*** disappear at $x, y \to \infty$ and Fourier transforms of ***all*** quantities (in particular displacements) exist. In the last expression for $\tilde{u}_z(0)$ in Eq. (A.14) $\tilde{T}(k, z_1)$ significantly differs from zero only at small k. In this region exp (- $k z$) ~ 1, and Fourier transform can be inverted:

$$l \equiv -u_z(0) = \frac{2b(1+s)}{3}\int_{0}^{\infty} T(z)dz\,. \qquad \text{(A.16)}$$

This result for surface displacement is valid ***independently*** on the spatial profile of the laser beam in 3D static elasticity if all stresses relax at infinity and heat conduction is 1D in the sense that spatial temperature distribution in *z*-direction is much smaller than in *x-y* direction. It is ***not equal*** to pure 1D case, when there is ***no*** stress relaxation at infinity.

Appendix B. Cleaning threshold with the single sinusoidal pulse

The solution of the problem Eqs. (69)-(71) ***during the pulse*** is

$$h_1 = \frac{lw^2(w\sin w_0 t - w_0 \sin wt)}{2pw_0(w^2 - w_0^2)}, \quad \dot{h}_1 = \frac{lw^3(\cos w_0 t - \cos wt)}{2p(w^2 - w_0^2)}. \qquad \text{(B.1)}$$

If particle detaches ***after*** the pulse, the total energy at the pulse end should be bigger than adhesion energy. In other words

$$\dot{h}_1^2(t) + w_0^2 h_1^2(t) > w_0^2 h_0^2 \Rightarrow \left| \frac{lw^3 \sin\frac{w_0 t}{2}}{p(w^2 - w_0^2)} \right| > w_0 h_0 \Rightarrow \frac{l}{h_0} > \left| \frac{py(1-y^2)}{\sin py} \right|. \qquad \text{(B.2)}$$

The situation is more complicated if the detachment occurs ***during*** the pulse. Turning points for h_1 are given by the condition

$$\dot{h}_1 = 0 \Rightarrow \cos ywt = \cos wt \Rightarrow ywt = 2pn \pm wt \Rightarrow wt = \frac{2pn}{y \pm 1}. \qquad \text{(B.3)}$$

where n is integer number. The value of h_1 at these turning points is:

$$h_1(\dot{h}_1 = 0) = \frac{l}{2p}\frac{\sin ywt - y\sin wt}{y(1-y^2)} = \frac{l}{2p}\frac{(\pm 1 - y)\sin wt}{y(1-y^2)} = \frac{l}{2p}\frac{\sin\left(\frac{2pn}{y \pm 1}\right)}{y(y \pm 1)}. \qquad \text{(B.4)}$$

Detachment during the pulse occurs if this expression is smaller than $-h_0$ for some n, in other words

$$\frac{l}{h_0} > \min_{n,\pm}\left(-\frac{2py(y\pm 1)}{\sin\left(\frac{2pn}{y \mp 1}\right)}\right). \qquad \text{(B.5)}$$

The argument of the sine function (which is always smaller than $2p$ as long as we are within the pulse) should be as close to $3p/2$ as possible (then the value of sine is close to - 1). It can also be shown, that lower sign should be always preferred (numerator is smaller for " - " sign, while denominator is always close to "- 1"), This results in the condition

$$n = \left[\frac{3}{4}(y+1)\right], \qquad \text{(B.6)}$$

where [] denotes the closest integer number. Finally, it can be verified that with $y < 1$ Eq. (B.2) is always smaller than Eq. (B.5), which means that with $y < 1$ particle always detaches after the pulse. For $y>1$ the situation is the opposite and particle always detaches during the pulse, at $w\,t \approx 3p / 2$ or $t/t \gg 3/4$, i.e., in the second half of the (symmetric) pulse. Combining these two cases, we arrive at the expression (73) in the text.

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