### Dry laser cleaning of particles from solid substrates: Experiments and theory

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The experimental analysis of dry laser cleaning efficiency is done for certified spherical particle  $(SiO_2, 5.0, 2.5, 1.0, and 0.5 \ \mu m)$  from different substrates (Si, Ge, and NiP). The influence of different options (laser wavelength, incident angle, substrate properties, i.e., type of material, surface roughness, etc.) on the cleaning efficiency is presented in addition to commonly analyzed options (cleaning efficiency versus laser fluence and particle size). Found laser cleaning efficiency demonstrates a great sensitivity to some of these options (e.g., laser wavelength, angle of incidence, etc.). Partially these effects can be explained within the frame of the microelectronics engineering (MIE) theory of scattering. Other effects (e.g., influence of roughness) can be explained along the more complex line, related to examination of the problem "particle on the surface" beyond the MIE theory. The theory of dry laser cleaning, based on one-dimensional thermal expansion of the substrate, demonstrates a great sensitivity of the cleaning efficiency on laser pulse shape. For the reasonable pulse shape this theory yields the threshold fluence by the order of magnitude larger than the experimental one. At the same time the theory, which takes into account the near-field optical enhancement and three-dimensional thermal expansion effects, yields the correct values for threshold. © 2001 American Institute of Physics. [DOI: 10.1063/1.1389477]

### I. INTRODUCTION

Recently, laser-particle-surface interaction has attracted much concern in experimental and theoretical studies. There are three main impetuses behind this problem: first, as the dimension of active elements shrinks drastically in integrated circuit (IC) and high-density hard disk manufacturing, even a submicron particle may induce fatal damages to the whole system. On the other hand, because the inertial force  $m\ddot{x}$ contains mass  $m \propto R^3$  and the adhesion force  $F_{adh} \propto R$ , the necessary acceleration is  $\ddot{x} \propto R^{-2}$  (*R* is radius of the particle).<sup>1</sup> Typically, it is about 10<sup>7</sup> times higher than the gravity force for a 1  $\mu$ m particle.<sup>2</sup> Traditional cleaning methods, such as hydrodynamic jet and ultrasonic vibration, cannot remove small particles efficiently. Fortunately, pulsed laser heating of the absorptive particle or substrate can produce such high acceleration.<sup>3-26</sup> Second, due to the simplicity of the laser-particle-surface system, it can be applied in the study of the particle adhesion and deformation on solid substrate, which is the basis of many cross-discipline subjects.<sup>27–31</sup> Third, the fascinating physics arises in the laser cleaning problem, the latest are, e.g., effects, related to nearfield focusing of radiation by particle with size comparable with laser wavelength.<sup>17-21</sup>

In recent studies of the dry cleaning mechanism, spherical particles of standard sizes are adapted in experiments. These particles, with almost uniform size and fine sphericity, can greatly minimize the uncertainties that are mostly induced by surface aspherities. From the viewpoint of practical application, as the sphericity of the particles tends to increase along with the reducing particle size,<sup>32</sup> it is necessary to explore the mechanism of laser cleaning for spherical particles from solid substrates. Furthermore, since most of the theoretical models are based on the simplification that particles are spherical, it is convenient to compare theoretical calculation and experimental results based on spherical particles.

However, most experimental investigations concentrated only on the influence of laser fluence and particle size on the cleaning efficiency. Other cleaning options, such as laser wavelength, incident angle, polarization, substrate properties, are not fully covered. In this article, the influence of these options on the cleaning efficiency is reported. The study of the influence of surface roughness, although preliminary due to the complexity of this topic, is also presented.

Another important topic is the interaction of laser light with the particle on surface, which is also ignored in previous studies. An exact solution of this problem shows that the near-field light intensity, produced by the optical resonance and substrate reflection, is wholly different from the intensity profile of the incident laser light. These calculation results can explain many experimental phenomena in laser cleaning, as presented in the following parts. The "true" light intensity profile should be applied in the cleaning model during the solution of heat equations (for calculating substrate thermal expansion and tensions).

### **II. EXPERIMENT**

In Secs. A–D, a KrF excimer laser (Lambda Physik) with a wavelength of 248 nm, pulse width of 23 ns, and a

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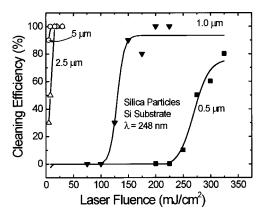


FIG. 1. Cleaning efficiency as a function of laser fluence for particles of different size. Size effect is obvious for small particles. However, the threshold laser fluence is not reversibly proportional to the square of particle size.

maximum pulse repetition rate of 30 Hz was used as the light source for laser cleaning. The fluctuation of fluence in the  $2.5 \text{ cm} \times 4.0 \text{ cm}$  spot is less than 7%. In Sec. E, the light source was Nd: yttrium-aluminum-garnet (YAG) laser (B. M. Industries 5000 Series). It consists mainly of a single *Q*-switched oscillator followed by an amplifier. Three detachable harmonic generators extend the wavelength range to the second, third, and fourth harmonics. The output light has a Gaussian distribution with a spot size of about 0.8 cm. The pulse duration is 7 ns.

# A. Laser cleaning of spherical particles from Si substrate

As in our previous article,<sup>21</sup> the particles of defined certified sizes (production of Duke Scientific Corporation) are used in the experiment. The particles are spherical and uniform in size. Figure 1 shows the cleaning efficiency as a function of laser fluence. It is found that the laser cleaning efficiency increases sharply along with the laser fluence. For particles with sizes of 0.5 and 1.0  $\mu$ m, threshold laser fluences are about 225 and 100 mJ/cm<sup>2</sup>, respectively. For 2.5 and 5.0  $\mu$ m particles, the threshold laser fluences are found to be lower than 5 mJ/cm<sup>2</sup>. This experiment shows that the size effect exists for small particles. However, it should be emphasized that the threshold fluences are not proportional

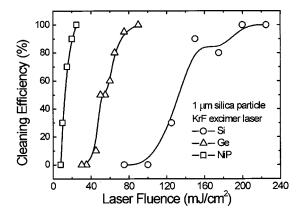


FIG. 2. Cleaning curves for NiP, Ge, and Si substrates. Total pulse number is 200.

to  $R^{-2}$ , this is because the rough estimation does not include the laser-particle interaction, which will be explored in Secs. III and V.

# B. Influence of substrate properties on cleaning efficiency properties

In this experiment, the different cleaning efficiencies of 1.0  $\mu$ m silica particle from Si, Ge, and NiP surfaces are examined, see Fig. 2. It is found that the laser cleaning efficiency increases sharply along with the laser fluence. For 1.0  $\mu$ m particles, threshold laser fluences exist at about 100 mJ/cm<sup>2</sup> for Si, about 30 mJ/cm<sup>2</sup> for Ge, and about 8 mJ/cm<sup>2</sup> for NiP substrate. It is also concluded that particle removal from NiP is the easiest in the three substrates, while removal from Si is the most difficult.

To analyze the differences of cleaning results, it is better to list the physical parameters of the materials, as shown in Table I. The van der Waals force of particles on Ge is the greatest of the three substrates, while the adhesion force on NiP is only about 2/3 of Ge. However, particle removal is related not only to the adhesion force, but also to the optical and thermal properties of the substrates. For Si, the absorptivity and thermal expansion coefficients are much less than Ge and NiP. Therefore higher laser fluence is required to remove particles from Si substrate. Removal from NiP is the

TABLE I. Parameters used in the calculations (taken at T = 300 K). Optical parameters are given for  $\lambda = 248$  nm (KrF excimer laser). Hamaker constant is given for the pair of identical materials, i.e., Si–Si, etc. For the pair of different materials *A* and *B* the Hamaker constant is approximated by  $A_{AB} = \sqrt{A_{AAB}}$ .

	Values for different materials						
Parameter	Si	Ge	NiP	SiO <sub>2</sub>			
Absorption coefficient (cm <sup>-1</sup> )	$1.8 \times 10^{6}$	$1.6 \times 10^{6}$	5.7×10 <sup>5</sup>	$5 \times 10^{2}$			
Absorptivity	0.33	0.35	0.71	0.95			
Density (g/cm <sup>3</sup> )	2.3	5.33	8.9	2.2			
Thermal conductivity (W/cm K)	1.42	0.73	0.14	0.0146			
Heat capacity (J/g K)	0.72	0.31	0.54	0.74			
Melting temperature (K)	1685	1210	1200	1873			
Young modulus (dynes/cm <sup>2</sup> )	$1.3 \times 10^{12}$	$8.2 \times 10^{11}$	$2.0 \times 10^{12}$	$7.3 \times 10^{11}$			
Poisson ratio	0.28	0.3	0.31	0.17			
Linear thermal expansion (K <sup>-1</sup> )	$2.6 \times 10^{-6}$	$6.0 \times 10^{-6}$	$12.0 \times 10^{-6}$	$0.54 \times 10^{-1}$			
Hamaker constant $A = 3\langle \hbar \omega \rangle / (J)$	$2.5 \times 10^{-19}$	$3.1 \times 10^{-19}$	$1.2 \times 10^{-19}$	$0.7 \times 10^{-1}$			

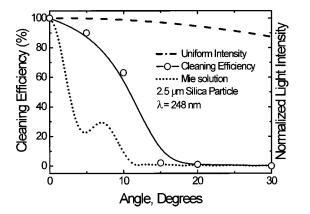


FIG. 3. A steep decline of cleaning efficiency appears with increasing incident angle. The uniform light intensity is multiplied by 100, the Mie solution is multiplied by 0.81. This figure shows that the near-field light intensity is sensitive to the incident angle.

easiest, no only because the adhesion force is the least of the three substrates, but also because its absorptivity and thermal expansion coefficients are much greater than the other substrates. The small thermal conductivity of NiP also contributes to its low threshold, since the real heating process is more analogous to "pointing heating" due to the near-field effect. This work indicates that substrate thermal expansion, enhanced by near-field optical focusing effects, is the dominant mechanism for dry laser cleaning.

#### C. Influence of incident angle on cleaning efficiency

To investigate the influence of incident angle, the Si pieces with particle contamination on the surface were placed on triangular planes that have tilting angles of  $5^{\circ}$ ,  $10^{\circ}$ ,  $15^{\circ}$ ,  $20^{\circ}$ ,  $30^{\circ}$ ,  $45^{\circ}$ , and  $60^{\circ}$ , respectively. Since the incident light was kept normal to the stage, the incident angles relative to the substrate surface were determined by the triangular planes. The surfaces before and after laser irradiation were observed with a high-resolution optical microscope.

The laser fluence is proportional to  $A(\theta)\cos\theta$ , where  $\theta$  is the incident angle, A is the absorptivity. The variation of incident angle from 0° to 15° caused the effective laser fluence to drop less than 3.5%. Since 43 mJ/cm<sup>2</sup> was far above the threshold laser fluence (below 5 mJ/cm<sup>2</sup> for 2.5  $\mu$ m particles), the particles should be completely removed, from the result in Fig. 1, with such a minus drop of fluence. However, the experiment shows that the cleaning efficiency drops to 0 when the incident angle is greater than 15° (Fig. 3). A similar result was found in Ref. 19. From the calculation based on the MIE theory, we found that the near-field light intensity declines from 100 to 0 when the tilting angle increases from 0° to 15°. This calculation is in agreement with the experimental results.

### D. Influence of surface roughness on cleaning efficiency

In the IC process, although the Si wafer starts with a perfect surface structure, the morphology will change drastically after a few steps of oxidation, etching, doping, diffusion, and mechanical polishing.<sup>33</sup> In this paragraph, the in-

fluence of surface roughness on the cleaning efficiency is investigated. The Si surfaces were modified with anisotropic etching in KOH solvent with different times, and subsequently observed by the atomic force microscope, as shown in Fig. 4. Surface roughness of samples is listed in Table II. It is found that both peak-valley roughness (Rp-v) and root mean square (rms) roughness increase with increasing etching time.

The cleaning curves for different samples are shown in Fig. 5. The particle size is 1.0  $\mu$ m. Although theoretical prediction shows that particle adhesion force drops obviously with the presence of very small roughness,<sup>34–36</sup> in this experiment, the increasing roughness produced less efficiency. In our early explanation, it is assumed that the surface elascity is reduced by the roughness. New examination shows that the near-field light intensity drops sharply with increasing particle-surface distance. Since the cleaning efficiency is sensitive to laser fluence, the reduction of the near-field light intensity, due to the surface asperities, subsequently causes less cleaning efficiency.

# E. Influence of laser wavelength on cleaning efficiency

In this section, *Q*-switched Nd:YAG laser with light wavelengths of 1064, 532, and 355 nm is applied as light sources to remove 0.5  $\mu$ m spherical silica particles from substrates. The cleaning curves are shown in Fig. 6. It is found that the cleaning effect for 1064 nm light is poor. The threshold laser fluence for a long wavelength is higher than that for short wavelengths.

The influence of wavelength on the cleaning efficiency can be ascribed to two aspects: First, the substrate absorption coefficient of laser light is different for different wavelengths. For Ge substrate, the absorption coefficients of 355, 532, and 1064 nm light are 0.5, 0.48, and 0.61, respectively, (at 300 K). Second, the interaction of laser light with particles results in localized light intensity near the particlesubstrate contacting area, which is highly related to the wavelength and particle size.

### III. INTERACTION OF PARTICLE AND LASER LIGHT: OPTICAL RESONANCE AND NEAR-FIELD EFFECT

The near-field light intensity is based on the solution of the boundary problem: a sphere particle on a flat semiinfinite substrate. The solution to this problem is rigorous, and it is beyond the scale of this article. The details can be achieved from reference  $^{23}$ .

For a nonabsorptive spherical particle, incident light could excite some resonance modes inside the particle and produce enhanced light intensities near the contacting area. In this case the particle acts like a lens, as was shown in Refs. 22–26.

Some laser cleaning phenomena in experiments can be explained by the near-field light profile. When incident light irradiates from a small tilting angle, the focusing point will shift away from the contacting point, therefore the cleaning efficiency drops rapidly. The cleaning efficiency is sensitive

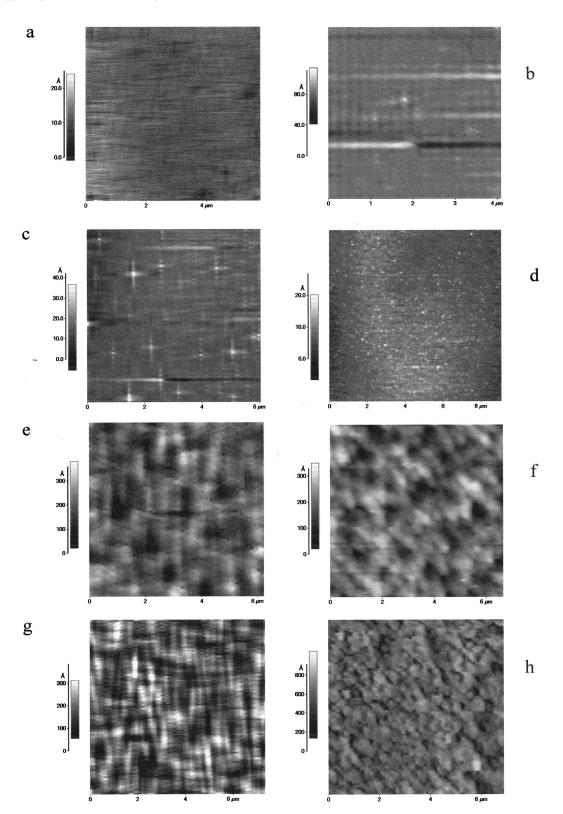


FIG. 4. Surface morphology with different etching times: (a) 0, (b) 1, (c) 5, (d) 10, (e) 15, (f) 20, (g) 25, and (h) 30 min, respectively. The roughness increases with etching time.

to the wavelength, because different wavelengths produce various intensity profiles. The near-field light intensity is also sensitive to the distance between particle and the rough surface, as we assume that the particle is "lifted up" by certain surface asperities. Figure 7 shows the surface light intensity drops sharply with increasing distance. The central light intensity reduces to almost half value of the case for particle on flat surface, as the distance reaches 50 nm. This distance is comparable to the roughness in experimental part D.

TABLE II. Surface roughness of different samples in Fig. 4.

Samples	a	b	c	d	e	f	g	h
Rp-v (nm)	2.28	1.35	1.45	1.54	31	34.7	40.1	63.8
Rms (nm)	0.137	0.207	0.143	0.188	4.68	5.52	6.06	9.06
Ave. rough (nm)	0.092	0.149	0.109	0.143	3.74	4.47	4.78	7.12

#### **IV. THEORETICAL MODEL**

Here a model for dry cleaning, which permits us to do an estimation of the threshold fluence for particle removal, is briefly discussed. Particles can be ejected from particulatecontaminated surfaces by short-pulse laser irradiation due to fast thermal expansion of the particle and/or solid surfaces. The following will focus on necessary steps in modeling of laser cleaning, which includes particle adhesion, thermal expansion of substrate, and criteria of particle removal.

### A. Adhesion energy and particle deformation induced by van der Waals force

The particle is attracted to the surface by van der Waals force, which occurs due to dipole interactions. If one considers the particle as a deformed sphere, then, according to Hamaker,<sup>37</sup> the attraction force is given by

$$F = \frac{\langle \hbar \, \omega \rangle R}{8 \, \pi h^2} + \frac{\langle \hbar \, \omega \rangle a^2}{8 \, \pi h^3},\tag{1}$$

where *R* is radius of the particle, *h* the separation distance  $(h \approx 4 \text{ Å})$ , and *a* the radius of contact. The Lifshitz constant  $\langle \hbar \omega \rangle$  is related to Hamaker constant, *A*, by  $A = 3/4 \pi \langle \hbar \omega \rangle$ . The Hamaker constant depends on the properties of the particle, substrate, and medium.

The attraction force (1) is very big; it is sufficient to say that the maximal pressure within the range of contact area consists, typically, of 10 Kbar or higher.<sup>38</sup> It is clear that this high loading leads to elastic or even plastic deformation of the material. Analyses of these deformations as well as the general problem of adherence are still under discussion, see, e.g., Ref. 39. The first, examination of pressure distribution within the contact area was done by H. Hertz in 1882; this distribution follows the parabolic law, see analysis of the

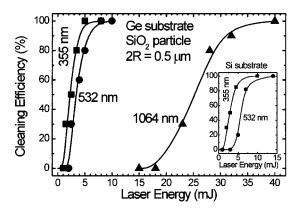


FIG. 6. Cleaning efficiency with respect to different wavelengths. Spot size is about  $0.3 \text{ cm}^2$ .

Hertz solution, for example, in Sec. 9 of the book.<sup>40</sup> Using this distribution, Derjaguin<sup>41</sup> found the relation between the radius of contact, a, and loading force,  $P_l$ , for spherical particle

$$a^{3} = \frac{3}{4} \frac{P_{l}R}{E^{*}}, \frac{1}{E^{*}} = \left(\frac{1 - \sigma_{p}^{2}}{E_{p}} + \frac{1 - \sigma_{s}^{2}}{E_{s}}\right), P_{\ell} = \frac{\langle \hbar \omega \rangle}{8\pi}, \quad (2)$$

where  $\sigma_{p,s}$  and  $E_{p,s}$  are the Poisson coefficients and Young's modulus for the particle (*p*) and substrate (*s*), the effective loading force, due to adhesion is presented by the first term in Eq. (1). The adhesion-induced deformations are quite complex, and some other factors (adhesion forces outside the area of contact, etc.) should be taken into account to describe well the experimental data. At present, two models of adhesion are commonly acceptable: Derjaguin–Muller–Toropov model for "hard" materials<sup>42,43</sup> and Johnson–Kendall– Roberts model for "soft" materials.<sup>44,45</sup> The transition between two models was also discussed.<sup>46,47</sup>

The distance between the center of the spherical particle and the surface of substrate for nondeformed materials is given by l = R + h. Due to deformation of materials under the action of external load and adhesion force, this distance will be l' < l. If no other load is exerted on the particle, the initial deformation parameter  $\delta_0$  is expressed in DMT theory as

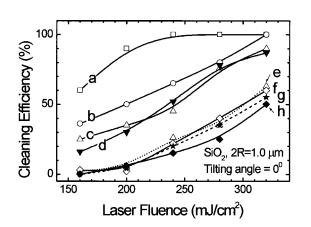


FIG. 5. Cleaning curves for different samples. The surface roughness increases with the alphabatic order. It is shown that higher roughness produces less efficiency.

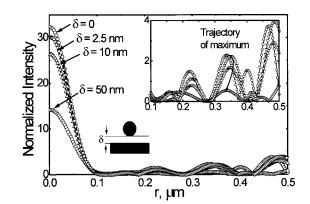


FIG. 7. Scattered light intensity on the substrate vs the distance between particle and the substrate. Light intensity drops drastically with increasing distance. The side-lobe structures first shift outward with the distance, then inward, reflecting the property of Newton rings.

$$\delta_0 = \frac{1}{8} \left[ \frac{9R \langle \hbar \, \omega \rangle^2}{2 \, \pi^2 h^4 E^{*2}} \right]^{1/3}. \tag{3}$$

The elastic repelling force  $F_e$  and the deformation energy  $W_e$  are expressed by

$$F_e = \frac{4}{3} E^* \sqrt{R \,\delta_0^3}, \quad W_e = \frac{8}{15} E^* \sqrt{R \,\delta_0^5}. \tag{4}$$

# B. Deformation and removal of silica particles during substrate surface expansion

When the substrate expands, position of the substrate surface,  $z_s$ , varies with time,  $z_s = z_s(t)$ ,  $z_s(0) = 0$ . The particle dislocation is a function of time *t*, say f(t). Then the running deformation parameter,  $\delta(t)$ , at time *t* can be expressed as

$$\delta(t) = z_s(t) + \Delta R(t) - f(t).$$
(5)

Here term  $\Delta R(t) = \alpha_T^{(p)} R T_s(t)$  describes the effect of the particle heating due to thermal contact; one considers that the temperature of the particle is the same as the substrate temperature.<sup>48</sup> ( $\alpha_T^{(p)}$ ) is the linear thermal expansion coefficient for the particle; we use an additional superscript to distinguish the particle and substrate materials. The acceleration due to the elastic force (4) can be expressed as<sup>10</sup>

$$\frac{4}{3}\pi R^{3}\rho_{p}\frac{d^{2}f(t)}{dt^{2}} = \frac{4}{3}\sqrt{R}E^{*}[\delta(t)^{3/2} - \delta_{0}^{3/2}],$$
(6)

where  $\rho_p$  is the density of the particle. The initial conditions for Eq. (6) are

$$\left. \frac{df}{dt} \right|_{t=0} = \frac{d\Delta R}{dt} \bigg|_{t=0}, \ f|_{t=0}.$$

$$\tag{7}$$

To find the surface position,  $z_s(t)$ , we use solution T = T(z,t) of the one-dimensional (1D) heat equation. From this solution one can estimate the surface displacement by

$$z_{s}(t) = 2(1+\sigma_{s}) \alpha_{T}^{(s)} \int_{0}^{\infty} T(z,t) d.$$
(8)

The additional multiplier  $2(1 + \sigma_s)$  in Eq. (8) appears when one considers the "quasi-1D approach," where displacements and stresses are considered to be equal to zero at infinity (at *x*-*y* plane). This term without detailed explanation was written in Refs. 49 and 50. The detailed examination of the thermal elasticity problem, which precisely introduces this term, is given in Ref. 51.

Because of the energy conservation, one can write condition

$$A \int_{0}^{t} I(t_{1}) dt_{1} = c \rho \int_{0}^{\infty} T(z, t) dz, \qquad (9)$$

where *c* and  $\rho$  are, respectively, heat capacity and density of the heated substrate, *A* is absorptivity of the surface. We consider the smooth pulse shape, typical for excimer laser pulse<sup>52</sup>

$$I(t) = I_0 \frac{t}{\tau} \exp\left[-\frac{t}{\tau}\right].$$
 (10)

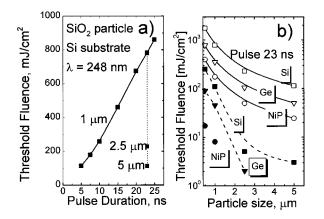


FIG. 8. (A) Threshold fluence for laser cleaning vs pulse duration for different size of particles. Two pulse shapes are examined: rectangular pulse and smooth excimer pulse. (B) Theoretical and experimental results of the threshold laser fluences for particles on Si, Ge, and NiP. Solid symbols are experimental data, while empty symbols are theoretical prediction based on 1D thermal expansion.

The laser fluence is given by  $\Phi = I_0 \tau$ , and pulse duration at the full width at half maximum  $t_{\text{FWHM}} \approx 2.446 \tau$ . The  $I_0$  is the near-field light intensity, which is different from the incident light.<sup>23</sup> From Eqs. (8)–(10) one can estimate the surface position,  $z_s(t)$ , by

$$z_s(t) = 2(1+\sigma_s) \frac{\alpha_T A \Phi}{c\rho} \bigg[ 1 - \bigg(1 + \frac{t}{\tau}\bigg) e^{-(t/\tau)} \bigg].$$
(11)

For the rectangular laser pulse with the pulse duration  $\tau_{\mathbb{Z}}$ , a similar dependence is given by

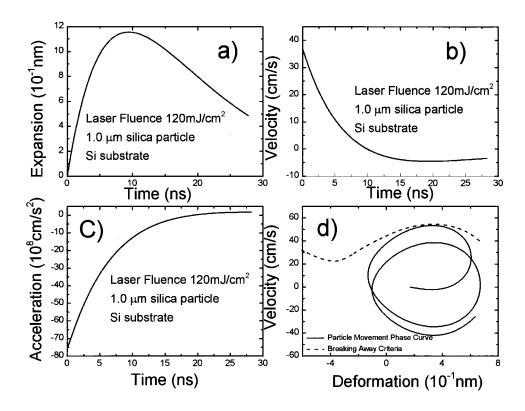
$$z_{s}(t) = 2(1+\sigma_{s}) \frac{\alpha_{T}A\Phi}{c\rho} \left[ \frac{t}{\tau_{\ell}} \Theta_{H} \left( 1 - \frac{t}{\tau_{\ell}} \right) + \Theta_{H} \left( \frac{t}{\tau_{\ell}} - 1 \right),$$
(12)

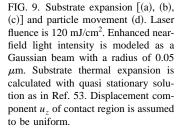
where  $\Theta_H(x)$  is the unit step function (the Heaviside function). The 1D model has natural restriction, related to absence of inward motion of the surface during cooling stage. To take this effect into account one should use the threedimensional (3D) thermo-elastic model.<sup>48</sup> Effects related to temperature dependencies of parameters (solution of nonlinear heat equation) can also play an important role. Nevertheless, because of its simplicity, the derived model can be used for the preliminary estimations.

Although the heating process happens only within a short time interval, it determines both the kinetic energy and the elastic potential energy necessary to overcome the adhesion force. The condition for the particle removal can be written from the energetic consideration<sup>10</sup> (see also Ref. 52 for the simplified energy criterion)

$$\frac{8}{15}E^*\sqrt{R\delta(t)^5} + \frac{4}{3}\pi R^3 \frac{\rho\nu^2}{2} \ge \frac{1}{2}P_l\delta(t) + \frac{\langle\hbar\omega\rangle R}{8\pi h}.$$
 (13)

In Fig. 8 one can see the result of the examination of the threshold fluence on the pulse duration and particle size. This picture presents the main tendencies in dry laser cleaning. First, the small particles are more difficult to remove than the big particles. Second, the threshold fluence is smaller for a





shorter laser pulse. Third, the cleaning effect strongly depends on the pulse shape. The smooth laser pulse (10), close to the true shape of excimer laser pulse,<sup>52</sup> yields significantly larger threshold fluence than those for rectangular pulse.<sup>21</sup> Nevertheless, even rectangular pulse predicts higher threshold fluences than the experimental one (see, e.g., Fig. 7 in Ref. 21). Thus, one should conclude that the theory based on 1D thermal expansion of the substrate<sup>1,7,10,17,21</sup> cannot explain experimental results on dry laser cleaning. Results of the recent articles<sup>15,19</sup> also do not confirm the 1D thermal expansion mechanism.

We suggested in Ref. 48 another mechanism of dry laser cleaning, based on the near-field optical enhancement effect. Due to this effect the transparent particle works as a focusing lens which produces on the substrate surface a "bright point," as it was confirmed experimentally.<sup>23,24</sup> The 3D thermal deformations of the substrate produce, in turn, sufficient accelerations to explain small threshold fluence.<sup>48</sup>

The equation for the displacement is given by the thermo-elasticity theory  $^{21,41}$ 

$$\rho \ddot{\mathbf{u}} = \frac{E}{2(1+\sigma)} \Delta \mathbf{u} + \frac{E}{2((1+\sigma)(1-2\sigma))} \operatorname{grad} \operatorname{div} \mathbf{u}$$
$$-\frac{\alpha_T E}{3(1-2\sigma)} \operatorname{gra}. \tag{14}$$

Solution of Eq. (14) for the stationary problem (laser beam with Gaussian intensity distribution) was presented in Ref. 53. Figure 9 is the calculation based on 3D "quasi stationary" thermal expansion. Figure 9(d) shows that the particle kinetic and elastic deformation energy is close to the breaking away criteria when laser fluence is 120 mJ/cm<sup>2</sup>, which is much less than the 1D threshold laser fluence. It is found that for displacements, e.g.,  $u_z$  component, this formal

solution in Ref. 53 contains logarithmic divergence, related to 1/r spatial distribution of the stationary thermal field at big distances.<sup>51</sup> It is clear that the corresponding stationary limit is senseless, i.e., in the real physical situation corresponding integrals should be cutoff. In the physically reliable region this "renormalization procedure" does not influence results strongly, nevertheless it is not strictly defined. For more precise analysis one should use the nonstationary solution in the "quasi-static" limit.<sup>51</sup>

### V. CONCLUSION

Although the mechanism of dry laser cleaning related to the thermal expansion of materials is commonly accepted, some details of this mechanism need clarification. The important point is related to the influence of particles on the distribution of laser intensity. The consequences of this influence were never precisely examined. In part, it is related to complexity of theoretical analysis for the problem particle on the surface. Our examination<sup>24</sup> shows, that for transparent spherical particles with the size of  $\sim\lambda$ , the optical resonance and substrate reflection produce "enhanced" near-field light intensity, which is sensitive to wavelength, particle size, incident angle, and surface morphology. These cleaning options are not fully covered in the known experiments.

Thus, the main goal of the present work was examination of the mentioned dependencies in experiments with strictly defined particle sizes. The physical model of laser cleaning, based on the idea of optical resonance and nearfield effects, predicts some effects which were attractive to confirm or disconfirm experimentally.

The results of the present work can be summarized as follows.

(1) The threshold fluence for laser cleaning strongly depends on the particle sizes (smaller particles need higher fluence). This dependence does not follow the law  $\Phi_{\rm th} \propto R^{-2}$  (see in Figs. 1 and 8).

(2) The strong angular dependence in the cleaning efficiency for small incident angles,  $\vartheta \leq 15^{\circ}$  (see Fig. 3) demonstrates that the intensity distribution, originated by near-field focusing effects, plays a major role in dry laser cleaning.

(3) The optical and thermal properties of materials play more important role in laser cleaning than the adhesion force by itself. Experiments (see in Fig. 2) show variations in threshold laser fluence by the order of magnitude for the materials where the Hamaker constants are practically the same. These results suggest that thermal expansion, enhanced by near-field optical focusing effects, is the dominant mechanism in dry laser cleaning.

(4) The nontrivial influence of the surface roughness was found in dry laser cleaning. It is well known that the surface roughness leads to a decrease in adhesion force. Thus, experiments with static load<sup>19–21</sup> demonstrate that the particle can be easily removed with the increase of roughness. At the same time calculation of the near-field effects shows that the laser intensity on the surface decreases rapidly with distance from the particle to the surface (see in Fig. 9). Thus, with increase of the surface roughness the efficiency of dry laser cleaning should rapidly decrease. Experiments (see Fig. 5) demonstrate the rapid decrease in the cleaning efficiency with growth of roughness.

Some interesting effects were, nevertheless, out of the frame of performed examinations. For example, with the particle size  $R \sim \lambda$ , the theory predicts the rapid variation in the cleaning efficiency with wavelength (for fixed size of particles) due to optical resonance.<sup>24</sup> To check this prediction one needs experiments with a tunable laser.

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- <sup>1</sup> W. Zapka, W. Ziemlich, and A. C. Tam, Appl. Phys. Lett. **58**, 2217 (1991).
- <sup>2</sup> Particles on the Surfaces I. Detection, Adhesion, and Removal, edited by K. L. Mittal (Plenum, New York, 1988).
- <sup>3</sup>S. J. Lee, K. Imen, and S. D. Allen, J. Appl. Phys. **74**, 7044 (1993).
- <sup>4</sup>J. D. Kelly and F. E. Hovis, Microelectron. Eng. 20, 159 (1993).
- <sup>5</sup>A. N. Jette and R. C. Benson, J. Appl. Phys. **75**, 3130 (1994).
- <sup>6</sup>R. Larciprette and E. Borsella, J. Electron Spectrosc. Relat. Phenom. **76**, 607 (1995).
- <sup>7</sup>Y. F. Lu, W. D. Song, M. H. Hong, T. C. Chong, and T. C. Low, Appl. Phys. A: Mater. Sci. Process. **64**, 573 (1997).
- <sup>8</sup>A. A. Kolomenskii, H. A. Schuessler, V. G. Mikhalevich, and A. A. Maznev, J. Appl. Phys. 84, 2404 (1998).
- <sup>9</sup>P. Liederer, J. Boneberg, M. Mosbacher, A. Schilling, and O. Yavas, Proc. SPIE **3274**, 68 (1998).
- <sup>10</sup> Y. F. Lu, Y. W. Zheng, and W. D. Song, Appl. Phys. A: Mater. Sci. Process. 68, 569 (1999).
- <sup>11</sup> V. Dobler, R. Oltra, J. P. Boquillon, M. Mosbacher, J. Boneberg, and P.

- <sup>12</sup>G. Vereecke, E. Röhr, and M. M. Heyns, J. Appl. Phys. 85, 3837 (1999).
- <sup>13</sup>X. Wu, E. Sacher, and M. Meunier, J. Appl. Phys. 86, 1744 (1999).
- <sup>14</sup>M. She, D. Kim, and C. P. Grigoropoulos, J. Appl. Phys. 86, 6519 (1999).
- <sup>15</sup>D. R. Halfpenny and D. M. Kane, J. Appl. Phys. 86, 6641 (1999).
- <sup>16</sup>R. Oltra, E. Arenholz, P. Leiderer, W. Kautek, C. Fotakis, M. Autric, C. Afonso, and P. Wazen, Proc. SPIE **3885**, 499 (2000).
- <sup>17</sup> Y. F. Lu, Y. W. Zheng, and W. D. Song, J. Appl. Phys. 87, 549 (2000).
- <sup>18</sup>X. Wu, E. Sacher, and M. Meunier, J. Appl. Phys. 87, 3618 (2000).
- <sup>19</sup>G. Vereecke, E. Röhr, and M. M. Heyns, Appl. Surf. Sci. **157**, 67 (2000).
   <sup>20</sup>D. R. Halfpenny, D. M. Kane, and R. N. Lamb, Appl. Phys. A: Mater. Sci.
- Process. **71**, 147 (2000). <sup>21</sup> Y. W. Zheng, Y. F. Lu, Z. H. Mai, and W. D. Song, Jpn. J. Appl. Phys., Part 1 **39**, 5894 (2000).
- <sup>22</sup> P. Leiderer, J. Boneberg, V. Dobler, M. Mosbacher, H.-J. Münzer, N. Chaoui, J. Siegel, J. Solis, C. N. Afonso, T. Fourrier, G. Schrems, and D. Bäuerle, Proc. SPIE **4065**, 249 (2000).
- <sup>23</sup> B. S. Luk'yanchuk, Y. W. Zheng, and Y. F. Lu, Proc. SPIE **4065**, 576 (2000).
- <sup>24</sup> M. Mosbacher, H.-J. Münzer, J. Zimmermann, J. Solis, J. Boneberg, and P. Leiderer: Appl. Phys. A: Mater. Sci. Process. (in press).
- <sup>25</sup> Y. F. Lu, L. Zhang, W. D. Song, Y. W. Zheng, and B. S. Luk'yanchuk, JETP Lett. **72**, 457 (2000).
- <sup>26</sup>H.-J. Münzer, M. Mosbacher, M. Bertsch, J. Zimmermann, P. Leiderer, and J. Boneberg, J. Microsc. (in press).
- <sup>27</sup>D. S. Rimai, L. P. DeMejo, and R. C. Bowen, J. Appl. Phys. 68, 6234 (1990).
- <sup>28</sup>A. Stalder and U. Durig, J. Vac. Sci. Technol. B 14, 1259 (1996).
- <sup>29</sup>L. J. Douglas and F. V. Swol, J. Chem. Phys. **106**, 3782 (1997).
- <sup>30</sup>A. B. Mann and J. B. Pethica, Appl. Phys. Lett. **69**, 907 (1996).
- <sup>31</sup>M. T. Bengisu and A. Akay, J. Acoust. Soc. Am. 105, 194 (1999).
- <sup>32</sup>K. L. Dishman, P. K. Doolin, and J. F. Joffman, Ind. Eng. Chem. Res. 32, 1457 (1993).
- <sup>33</sup>G. Y. Chang and S. M. Sze, ULSI Technology (McGraw-Hill International, Singapore, 1996).
- <sup>34</sup> D. J. Whitehouse and J. F. Archard, Proc. R. Soc. London, Ser. A **310**, 97 (1970).
- <sup>35</sup>D. M. Schaefer, M. Carpenter, B. Gady, R. Reifenberger, L. P. Demejo, and D. S. Rimai, J. Adhes. Sci. Technol. 9, 1049 (1995).
- <sup>36</sup> M. Soltani, G. Ahmadi, R. G. Bayer, and M. A. Gaynes, J. Adhes. Sci. Technol. 9, 453 (1995).
- <sup>37</sup>H. C. Hamaker, Physica (Amsterdam) **4**, 1058 (1937).
- <sup>38</sup>R. A. Bowling, in Ref. 2, p. 77.
- <sup>39</sup> Fundamentals of Adhesion and Interfaces, edited by D. S. Rimai, L. P. DeMejio, and K. L. Mittal (VSP, Utrecht, Netherlands, 1995).
- <sup>40</sup>L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon, New York, 1975).
- <sup>41</sup>B. V. Derjaguin, Kolloid-Z. 69, 155 (1934).
- <sup>42</sup>B. V. Derjaguin, V. M. Muller, and Yu. P. Toporov, J. Colloid Interface Sci. 53, 314 (1975); 73, 293 (1980).
- <sup>43</sup> V. M. Muller, V. S. Yushchenko, and B. V. Derjaguin, J. Colloid Interface Sci. **77**, 91 (1980); **92**, 92 (1983).
- <sup>44</sup> K. L. Johnson, K. Kendall, and A. D. Roberts, Proc. R. Soc. London, Ser. A **324**, 301 (1971).
- <sup>45</sup>K. L. Johnson, in *Theoretical and Applied Mechanics*, edited by W. T. Koiter (North-Holland, 1976), p. 133.
- <sup>46</sup>D. Maugis, J. Colloid Interface Sci. **150**, 243 (1992).
- <sup>47</sup> D. Maugis and B. Gauthier-Manuel, in *Abstracts of Papers of the American Chemical Society, Part 2* (American Chemical Society, Washington, DC 1993), p. 206.
- <sup>48</sup> B. S. Luk'yanchuk, Y. W. Zheng, and Y. F. Lu, The International Symposium on Intensive Laser Action and its Applications, August 21–25, 2000, Saint Petersburg, Russia. To be published in Proc. SPIE (2001).
- <sup>49</sup>A. M. Prokhorov, V. I. Konov, I. Ursu, and I. N. Mihailescu, *Laser Heat*ing of Metals (Hilger, Bristol, 1990).
- <sup>50</sup> M. Vicanek, A. Rosch, F. Piron, and G. Simon, Appl. Phys. A: Mater. Sci. Process. **59**, 407 (1994).
- <sup>51</sup>N. Arnold, G. Schrems, T. Mühlberger, and D. Bäuerle (to be published).
- <sup>52</sup>D. Bäuerle, Laser Processing and Chemistry, 3rd ed. (Springer, Berlin, 2000).
- <sup>53</sup>L. P. Welsh, J. A. Tuchman, and I. P. Herman, J. Appl. Phys. **64**, 6274 (1988).