First publ. in: Applied Physics / A, Materials Science and Processing, 72 (2001), 1, pp. 41-44

Optical field enhancement effects in laser-assisted particle removal

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Received: 31 May 2000/Accepted: 7 September 2000/Published online: 22 November 2000 – © Springer-Verlag 2000

Abstract. We report on the role of local optical field enhancement in the neighborhood of particles during dry laser cleaning (DLC) of silicon wafer surfaces. Samples covered with spherical colloidal particles (PS, SiO_2) and arbitrarily shaped Al_2O_3 particles with diameters from 320–1700 nm were cleaned using laser pulses with durations from 150 fs to 6.5 ns and wavelengths ranging from 400–800 nm. Cleaned areas were investigated with scanning electron and atomic force microscopy. Holes in the substrate with diameters of 200–400 nm and depths of 10–80 nm, depending on the irradiation conditions, were found at the former positions of the particles. For all pulse durations analyzed (fs, ps, ns), holes are created at laser fluences as small as the threshold fluence. Calculations of the optical field intensities in the particles' neighbourhood by applying Mie theory suggest that enhancement of the incident laser intensity in the near field of the particles is responsible for these effects. DLC for sub-ns pulses seems to be governed by the local ablation of the substrate rather than by surface acceleration.

PACS: 81.65.C; 79.60.Bm

Submicron particles on Si wafers are responsible for production losses or malfunctions in microelectronic materials unless removed [1]. Recently a cleaning strategy called dry laser cleaning (DLC) has proven to be efficient for the removal of particles with diameters down to 100 nm [2–10]. This technique is based on the irradiation of the substrate with a short laser pulse. It is assumed that thermal expansion of the substrate surface caused by the absorption of the laser energy plays the major role in the cleaning mechanism. This thermal expansion is thought to accelerate the particles, which are subsequently detached due to their inertia.

Concerning the industrial applications of this technique, an obvious requirement is to avoid surface damage induced by the cleaning process itself. Some authors have reported the appearance of particle-induced damage of the whole irradiated surface area during DLC of irregularly shaped Al_2O_3 particles from glass [7, 11]. For spherical particles it is known [12] that they may act as spherical lenses and therefore increase the laser intensity if their diameter is bigger than the laser wavelength $d \geq \lambda$. For $d \leq \lambda$ field enhancement at the particles is expected according to Mie theory. As submicron spherical particles are frequently used as model contaminants in laser cleaning studies [6, 8–10, 13–15], we investigate systematically the consequences of local field enhancement on DLC.

1 Mie calculation of field intensities

Calculations based on the Mie scattering theory using a modified version of a program described in [16] were performed to compute the optical field enhancement at dielectric polystyrene spheres ($n = 1.6$). As a first approach we neglected the influence of the silicon surface and performed the calculations for a particle in free space. For the spherical particles and wavelengths used in our experiments (Mie parameter $\pi d/\lambda$ from 1.3 to 10.1) we computed enhancements of the incident laser intensity of 2 ($d = 320$ nm) to 30 ($d = 1700$ nm). In general the intensity enhancement increases with increasing d/λ. A typical spatial distribution of the laser intensity at a PS sphere ($d = 800$ nm, $\lambda = 800$ nm) is shown in Fig. 1. The laser intensity has been plotted over a cross section parallel to the incoming laser beam and along the surface plane of an imaginary substrate located under the particle (for the influence of a real surface see Sect. 4). For this example at the light-averted side of the particle the laser intensity exhibits an enhancement by a factor of 11 compared to the incident intensity, with its maximum localized to an area some hundred nanometers in diameter.

These calculations indicate that not only spheres with a diameter $d \geq \lambda$ focus the incoming light, as expected from geometrical optics and demonstrated in [12], but also those with $d \leq \lambda$. This effect has recently been demonstrated experimentally [17]. Bearing in mind DLC applications, these results also imply that the actual laser intensities at the particle location can be considerably higher than the nominal ones applied. Such a phenomenon might have important consequences for the interpretation of laser cleaning experiments

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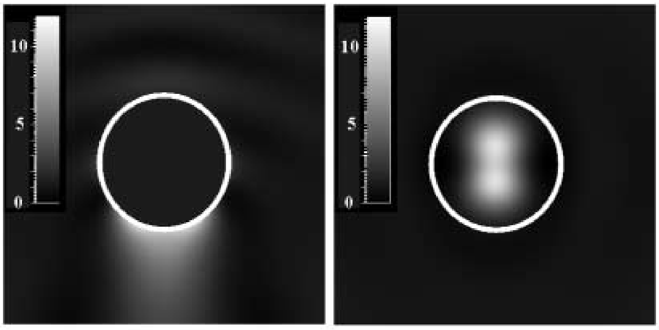

Fig. 1. Calculated laser intensity for a PS sphere ($n = 1.6$, $d = 800$ nm) illuminated with a plane electromagnetic wave at a wavelength of $\lambda = 800$ nm. Cross sections as seen from the side (*left*) and in the imaginary plane of the substrate surface as seen from above (*right*) are shown. The light field enters from the top, and the polarization vector of the electric field is indicated by an *arrow*. The displayed area is 2400×2400 nm^2. Note that in the left image the field inside the sphere was not calculated and the sphere is shaded grey for illustration only

where the laser fluences specified correspond to the average over the irradiated surface, especially for the fluence values quoted for the cleaning and damage thresholds. Additionally, as very high intensities are actually applied at the particle location, the risk of surface damage is very high, as will be shown in the following section.

2 Experimental details

Monodisperse polystyrene (PS) (Interfacial Dynamics, Portland, Ore., USA) or silica (SiO_2) spheres (Duke Scientific, Palo Alto, Calif., USA; Bangs Laboratories, Fishers, Ill., USA) with diameters in the range of 320 nm to 1700 nm and irregularly shaped alumina particles (Summit Chemicals Europe, Düsseldorf, Germany) were applied to the sample by spin coating after the particle suspension had been rarefied with isopropyl alcohol (IPA) [13–15]. Both materials are transparent at the wavelengths used.

Particle removal was carried out by irradiating the surface in air with laser pulses of different duration and wavelength. The fs pulses (FWHM = 150 fs) were generated by a mode-locked Ti:sapphire oscillator (Tsunami, Spectra Physics) coupled to a regenerative amplifier pumped by a 10 Hz Nd:YAG laser (TSA, Spectra Physics). For our experiments we used the fundamental and frequency-doubled wavelengths (800 nm/400 nm) of the system. A pulsed dye amplifier seeded either with a cw dye laser beam or with a synchronously pumped mode-locked dye laser [18] ($\lambda = 583$ nm) was also used for generation of both ns (FWHM = 2.5 ns) and ps (FWHM = 30 ps) pulses. For the generation of ns pulses we used a frequency-doubled Q-switched Nd:YAG laser ($\lambda = 532$ nm, FWHM = 6.5 ns). An OPO pumped by this Nd:YAG laser provided pulses at $\lambda = 800$ nm that enabled the comparison of the ns and fs regimes at the same wavelength.

We determined the laser fluences in the following way: the beam profile of the fs laser pulse was imaged on a CCD camera, and the total pulse energy was detected for each pulse. A combination of the results provided a spatially resolved mapping of laser fluences. For the ns pulses, determination of laser fluences is described in [15].

3 Local optical field enhancement induced surface damage

A comparison of the calculated local laser intensity enhancement with the difference between the cleaning and damage thresholds of the bare surface in the laser cleaning process (2 to 10, depending on the cleaning wavelength and laser pulse duration [13–15, 19]) strongly suggests that there is a great risk of damaging the substrate underneath the particles, even though the nominal laser fluence applied is below the substrate damage threshold.

3.1 Cleaning with fs and ps pulses

Scanning electron microscope (SEM) examination of the DLC cleaned regions showed the presence of holes all over the area for both ps and fs pulses. An example of this can be seen in Fig. 2, showing a sample contaminated with 800 nm PS particles cleaned with ps pulses ($\lambda = 583$ nm, FWHM = 30 ps). A particle as well as three holes can be seen. Atomic force microscopy inspection of these holes revealed typical depths of 70 nm and diameters of 250 nm.

In order to prove that the appearance of surface damage is related to the local field enhancement caused by the particles, we prepared samples with higher particle densities to allow formation of particle dimers, trimers, etc., and additionally marked the initial particle positions. This was done by evaporating a 10 nm SiO_2 layer onto the sample. After removal of the particles their size and position could be determined by local contrast changes in the SEM due to the different thickness of the oxide layers at and outside the former particle site. By a comparison of the hole diameters and morphologies with and without the applied oxide layer, we checked that this layer did not influence the optical properties of the particles.

An example of such a marked sample is shown in Fig. 3a. The original particle locations can be identified by the faint

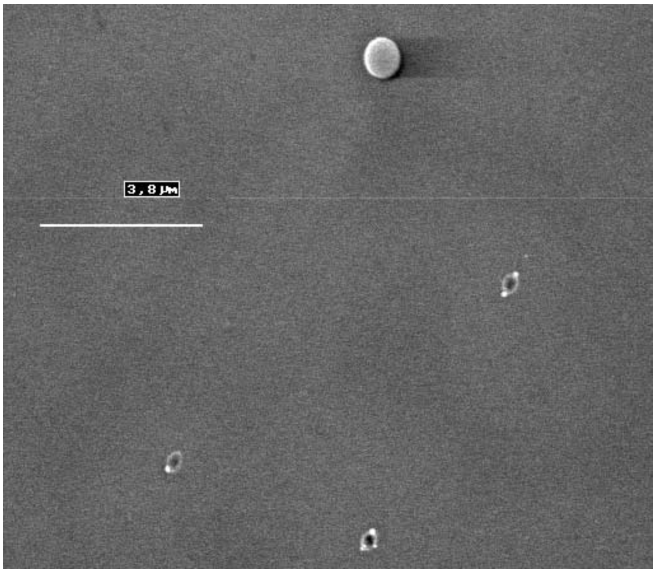

Fig. 2. Scanning electron micrograph of a cleaned area (DLC, FWHM = 30 ps, $\lambda = 583$ nm) in a sample contaminated with 800 nm PS spheres. Elliptical holes can be found all over the illuminated region; a particle can also be seen

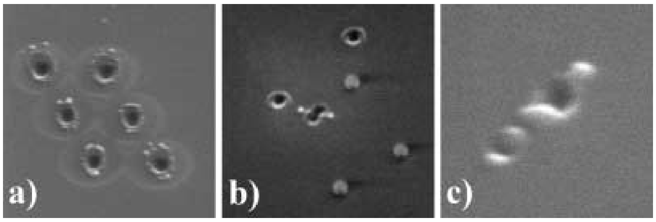

Fig. 3a–c. SEM micrograph of Si surfaces showing the holes generated during DLC ($\lambda = 800$ nm, FWHM = 150 fs). In (**a**) and (**b**) the particles are PS spheres with a diameter of 1700 and 320 nm, respectively; in (**c**), irregular alumina particles with a size of 400 nm are shown. The size of each figure corresponds to a square with a side of 6800 nm, 4970 nm, and 1300 nm for a, b and c, respectively

circular rims (diameter 1700 nm). Under each former particle a hole was created at the center of the marked area, where the numerical calculations predicted the highest intensity. Also for smaller particles holes at the former position could be found, as shown in Fig. 3b for spheres 320 nm in diameter. Note that the diameters of the particles used cover the range of 1700–320 nm and that the cleaning wavelength was 800 nm. From the ratio of d/λ it is evident that the enhancement of the laser intensity cannot properly be described by geometrical optics. Focusing in the near field of the particle as computed in Sect. 1 has to be taken into account. In general, the morphology of holes created by fs and ps pulses at $\lambda = 800$ nm, $\lambda = 583$ nm as well as $\lambda = 400$ nm was very similar. Typical diameters as measured by AFM were 200–400 nm and typical depths some 10 nm.

Important for the application of the DLC is the determination of the laser fluence threshold necessary for creation of such holes, in the following denoted as the "hole threshold". Figure 4 shows the edge of a region cleaned in ambient conditions by a fs pulse. Due to the continuous decrease in laser fluence from the beam center to the edge of the irradiated area, the border of the cleaned region marks a specific laser fluence. As can be seen from the image, even the minimum laser fluence necessary for removal of 800 nm PS spheres (marked by the edge of the cleaned area) led to the creation of holes. After dissolving the particles in toluene, we inspected the same area again and checked that there were no holes under particles that were not removed by the laser pulse. The same observation was made for 1700 nm and 320 nm PS spheres. Thus the hole threshold is equal to the cleaning threshold for these pulse durations and particle sizes, and it can be determined by measuring the threshold for particle removal. This threshold was determined from images of cleaned areas and the spatial mappings of laser fluences as described in Sect. 2. For the laser parameters $\lambda = 800$ nm and FWHM = 150 fs the obtained values for the incident, not enhanced laser beam were 11 $\mathrm{mJ/cm^2}$ for PS spheres 1700 nm in diameter, 25 $\mathrm{mJ/cm^2}$ for 800 nm spheres and 80 $\mathrm{mJ/cm^2}$ for 320 nm particles.

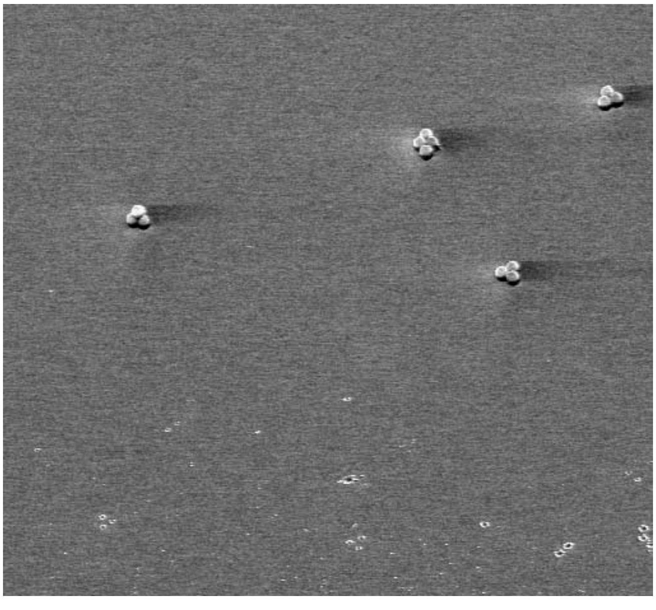

Fig. 4. The edge of a cleaned region after DLC of 800 nm PS spheres. Holes are created at fluences as small as the cleaning threshold laser fluence

3.2 Cleaning with ns pulses

For the case of ns pulses (FWHM = 6.5 ns) the thermal diffusion length during the pulse is on the order of several hundreds of nanometers, considerably larger than for the ps and fs case (several tens of nm). Therefore here heat diffusion cannot be neglected and influences the shape of the laser-induced holes. Typical examples of holes created at $\lambda = 800$ nm and FWHM = 6.5 ns are shown in Fig. 5a. When compared to those formed upon illumination with fs and ps pulses their size is larger and their depth is smaller. In addition to the holes, another damage surface structure could be found with a saucer-like shape, especially at lower laser fluences. A typical SEM picture of these structures is shown in Fig. 5b. For these lower laser fluences approaching the cleaning threshold, the damage structures become smaller, but can still be detected at the cleaning threshold for PS spheres with diameters approximately equivalent to the wavelength.

3.3 Particles with nonspherical shape

Realistic contaminants that have to be removed by laser cleaning applications are mostly of nonspherical shape. Spherical colloidal particles are used only as model contaminants due to their advantageous, well-characterized shape

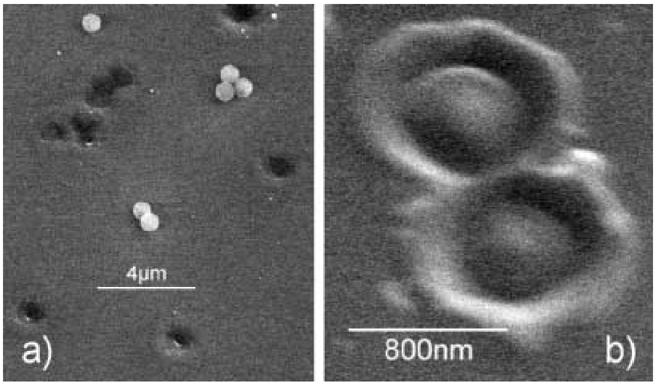

Fig. 5a,b. SEM micrograph showing the holes generated in the substrate after removal of 800 nm PS spheres with laser pulses at $\lambda = 800$ nm, FWHM = 6.5 ns. **a** For the higher laser fluences holes are found; **b** at lower fluences saucer-like structures with a small depth are seen and size. Therefore it should be investigated whether particles with irregular geometry induce effects similar to those described above. As contaminants we chose in this case 400 nm Al_2O_3 particles (Summit Chemicals Europe, Düsseldorf, Germany) that were deposited as predominantly isolated particles on the surface. After laser-assisted removal of these particles using fs pulses at $\lambda = 800$ nm, we also found holes, as depicted in Fig. 3c. This shows that local field enhancement also takes place at irregularly shaped particles. The fact that holes were not created at all particle locations may be explained by the random orientation of the irregularly shaped particles on the surface. Depending on the individual orientation, the field enhancement will be different for each particle and exhibit an asymmetric pattern, leading to the formation of holes with different shapes and sizes (see Fig. 3c). In some cases the enhancement might even be too small to create a hole.

4 Discussion and conclusion

In our study we were able to demonstrate the occurrence of local field enhancement at spherical and irregularly shaped particles irradiated by laser pulses ($\lambda = 532$–800 nm, FWHM $= 150$ fs to 6.5 ns). The existence of such local intensity enhancement is already indicated by the calculations presented in Sect. 1. They neglect the influence of the surface and therefore only provide a first estimation. However, very recently [20] it has been shown by numerical simulations that the local intensity enhancement in the presence of an Si surface might even be higher than calculated for a particle in free space.

Recently several models [6, 8, 9] have been proposed for the description of DLC. None of these models incorporates the effect of field enhancement; only in [8] is the possibility of “micro focusing” by the particles (size ≥ 500 nm, $\lambda = 248$ nm) mentioned, but it is not incorporated quantitatively. Our results suggest that modelling of the process needs to take into account the *enhanced intensities* underneath the particles. This is further illustrated by a comparison of the hole threshold for 800 nm particles (25 mJ/cm^2) with the ablation threshold for a bare surface at the same laser parameters of about 200–250 mJ/cm^2 [21]. Although the nominal intensity is far below the ablation threshold, the intensity underneath the particle that is enhanced by a factor of about 10 is in the range of ablation; hence holes are created.

In the aforementioned models it was proposed that the major mechanism responsible for particle detachment is the thermal expansion of the substrate surface and/or the particles due to heat absorption and the resulting inertia forces. We were able to identify another cleaning mechanism valid especially for ps and fs pulses that has not been reported in the literature so far. From the coincidence of the hole threshold and cleaning threshold fluences, we conclude that the particles in this case were removed by local ablation of the silicon substrate rather than by inertia forces. This local ablation mechanism dominates DLC for sub-ns pulses, probably due to the small thermal diffusion length and nonlinear effects (multi-photon absorption, avalanche breakdown) in the Si.

As well as DLC there is a another cleaning strategy, called “steam laser cleaning” (SLC) [2, 3], under investigation. It relies on the explosive evaporation of a water/alcohol mixture condensed onto the contaminated surface after the surface is heated by the laser pulse. In our previous studies we could show that SLC is more efficient than DLC [19] and provides an universal cleaning threshold for particle sizes from 60 to 800 nm [15]. With respect to surface damage by hole creation, preliminary experiments at $\lambda = 532$ nm, FWHM $= 8$ ns show another advantage. Due to the reduction of the field enhancement by reducing the refractive index difference between particle and ambient via application of the liquid, there exists a “safe” interval in laser fluence in between the cleaning threshold (110 mJ/cm^2, [15]) and the damage threshold of a bare Si surface (280 mJ/cm^2) where SLC without substrate damage is possible. Another form of SLC relies on the evaporation of the liquid by a laser (e.g. CO_2, $\lambda = 10.6\ \mu$m) that is absorbed in the liquid rather than in the sample [22]. Because in this case the wavelength is much larger than typical particle diameters, field enhancement induced damage should be reduced.

Our results have important consequences for possible applications of laser cleaning methods. The observation of hole formation does not mean that laser cleaning is not applicable to real world wafer processing, but it shows that cleaning methods in which field enhancement is reduced (e.g. SLC) should be considered.

Acknowledgements. This work was supported by the Optikzentrum Konstanz and the EU (TMR ERB-CT98-0188 “Modeling and diagnostic of pulsed laser–solid interactions: applications to laser cleaning”).

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