The power of light: Self-organized formation of macroscopic amounts of silica melts controlled by laser light

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CO_2 laser systems with a power output of up to 12 kW continuous wave have been employed for melting high purity amorphous silica (SiO_2) powders. Under the intense light irradiation, the migration of matter on the silica sample has been observed. A net mass transport results in the formation of macroscopic structures in the liquid phase. Protrusions of up to 7 mm height are formed against gravitational force and surface tension. For the first time, this work reports on the self-organized formation of macroscopic structures by viscous flow of a dielectric melt driven by laser light. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3000637]

Light-matter interaction can be described by the two basic mechanisms: reflection and absorption. The portion of the light, which is neither absorbed nor reflected, is transmitted. Absorption of light requires dissipation mechanisms, such as photon-phonon or exciton-phonon interactions. A more subtle type of the light-matter interaction can be observed when microscopic objects are irradiated by intense light. Impinging photons can transfer their momentum to the object exposed to light. This phenomenon, designated as radiation pressure, is relevant in the case of reflection and absorption. In the case of transparent objects, forces can originate from the electric-field gradient established by the light. With a focused laser beam, considerably strong fields can be established stable over time in a well defined volume. This phenomenon finds application in the so-called optical tweezers technology.[1–3] Although the typical magnitude of the applied forces is in the range of a few piconewtons, the manipulation of microscopic objects by light has evolved from simple experimental setups to sophisticated devices which have become powerful tools in physics as well as in biology.[4–6] A wealth of literature is devoted to this subject. However, to the best of our knowledge, the manipulation of macroscopic amounts of matter by light has not been reported yet. The present experiment will show, for the first time, that light induced forces can result in the manipulation of indeed macroscopic amounts of matter. As the major concern of the present paper is the introduction of this phenomenon, the experimental setup will be introduced and results will be discussed on a qualitative base.

Recently we have introduced the sintering of ultrapure silica (SiO_2) powder compacts by high power CO_2 laser systems.[7,8] The laser light emitted by CO_2 lasers has typically a wavelength of 10.6 μm. Light with this wavelength is readily absorbed by most ceramics via direct photon-phonon coupling.

For the present experiment we have employed a multimode CO_2 laser system from Trumpf Laser- und Systemtechnik GmbH, Germany (type TLF 12000 turbo), providing a power output of up to 12 kW continuous wave (cw). The linear polarized primary laser beam is shaped by a single convex mirror optics with a focal length of $f=300$ mm. The sample is placed relative to the optics such that a circular laser spot of 100 mm diameter is generated on the sample. The laser beam impinges with normal incidence to the horizontally oriented sample surface. With the given parameters a maximum mean power density of 154 W/cm^2 is calculated on the sample. With the given power density, temperatures exceeding the melting point of amorphous silica, that is, 1713 °C,[9] could be reached. The sample consists of a high purity SiO_2 powder (sum of all contamination is below 1 ppm) purchased from Mitsubishi Co., Japan, with a mean particle size of $d_{50}=40$ μm. In the present work, the laser treatment of the pure powders and powder compacts will be compared. Powder compacts are prepared by means of pressure casting. In a especially designed pressure casting process, providing high purity compacts, no organic additives have to be added for obtaining mechanically stable compacts. High purity fumed silica has been used as an inorganic binder.[8,10] The content of fumed silica in a powder compact amounts up to 10 wt %.

In laser treatment of SiO_2 powder samples, due to the low penetration depth of the laser light in the SiO_2, the light energy dissipates into heat at the powder samples surface. At maximum laser power output, i.e., a mean power density of 154 W/cm^2, the laser treated surface melts almost instantaneously. In order to avoid excessive evaporation of silica, the laser annealing experiments have been performed at a mean power density of 123 W/cm^2, corresponding to a laser power output of 9.6 kW. A prolonged laser treatment results in the formation of a pool of molten silica. Because of the poor heat conductivity within the powder, the depth of the melt pool increases relatively slow. The typical duration of a laser treatment ranges from 30 up to 300 s. At maximum duration, a melt pool of about 7 mm depth can be formed. At the beginning of the laser treatment a layer of partially molten silica particles mixed with their melt is formed at the substrate surface. In this stage we have observed the migration of significant amounts of material along and perpendicular to the surface. Protrusions of a height of up to 7 mm are formed at certain areas of the laser irradiated surface. Figure

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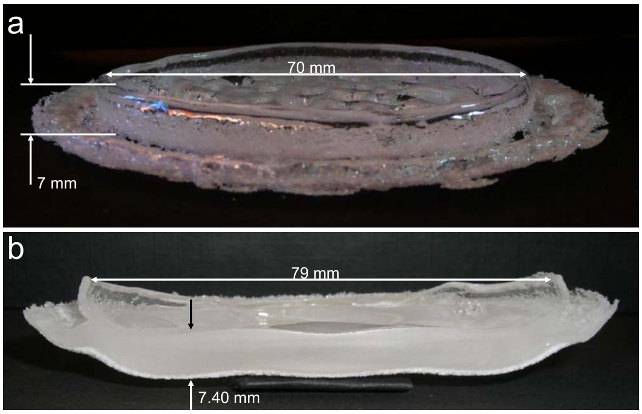

FIG. 1. (Color online) Quartz glass structure formed on an initially flat SiO_2 powder substrate after a (a) 95 s and (b) 300 s laser anneal at a mean power density of 123 W/cm^2, corresponding to a laser power output of 9.6 kW. The sample structure established during laser annealing is preserved by quenching the samples temperature after laser annealing by simply turning off the laser system. State of picture (b) presents a cross section of the sample.

1 presents the molten and resolidified part of a SiO_2 powder sample after annealing for (a) 95 s and (b) 300 s, respectively. Subsequent to the annealing phase the sample temperature is quenched by turning off the laser. Thus the structure formed in the liquid phase is preserved. Figure 1 basically reflects the original orientation of the sample in the experiment. In order to homogenize the radial temperature distribution within the laser spot, during the experiment the sample has been rotated at a speed of 70 rpm around the axis of the beam. However, similar results are obtained with the sample that is not been rotated. On the samples clearly noticeable is a ringlike protrusion of completely molten and resolidified, which is transparent, material. The protrusion of the surface appears in a circular zone of the highest intensity.

For laser treatment of SiO_2 powder compacts, in order to gain more insights into the mechanisms governing the formation of the protrusions during melting of silica powder samples, we have done an experiment similar to the above described, however, with silica powder compacts as a sample. In contrast to the sample made of loose silica powder, the powder particles in a compact are adhesively linked to each other by van der Waals forces. Therefore a powder compact provides mechanical strength to a certain degree already before sintering. By applying the same laser treatment to a silica compact we have not been able to generate a structure similar to the one shown in Fig. 1. No significant migration of material has been observed in the initial stages of laser treatment. However, after a prolonged laser treatment we found subsequent to the resolidification of an approximately 7 mm deep silica melt pool a surface topology, which virtually is an image of the intensity distribution within the laser spot. From the fact that an overheated silica melt can evaporate relatively quickly,[11] we have expected a loss of matter at highest intensity areas. In contrary, we found that the high intensity areas are elevated relative to those exposed to lower intensities. This observation strongly suggests that the liquid SiO_2 tends to migrate toward areas with relatively higher light intensity.

On the basis of these observations, we conclude that the interaction of a silica surface with intense laser light does not only merely result in the dissipation of the radiation energy into heat but also has a component in which the laser light applies forces to the melt. The existence of these forces is manifested by the formation of surface protrusions against gravitational force and surface tension. However, the formation of protrusions of the type presented in Fig. 1 is observed on powder samples only. More precisely, in the initial stage of the laser treatment of powder samples, when at the surface of the sample partially molten SiO_2 particles coexist with their melt. Once the protrusions are formed, they survive a prolonged laser treatment. On the other hand, on powder compacts no protrusions are formed even after excessive laser treatment when a pool of molten SiO_2 would provide sufficient mobility. In the following we suggest a model in which the surface tension of the sample is a determinant factor in the discussed scenario.

It is well established that light can apply forces to dielectric media. For the manipulation of microscopic objects as small as molecules, focussed laser beams are employed as the so-called optical tweezers. Light induced forces felt by a microscopic object trapped by a focussed laser beam consist of the light scattering and gradient forces and are in the range of a few piconewtons. In the Rayleigh regime, where the object diameter is relatively smaller than the wavelength of the light, forces are described through electromagnetic theory, see for a review Ref. 12. The object is treated as an induced point dipole, while its volume and geometry are neglected. This gradient force points in the direction of the intensity gradient of the light, while the scattering force points in the direction of the incident light. In our experiment we have annealed the surface of an amorphous SiO_2 powder with a grain size of $d_{50}=40$ μm. As the individual grains are significantly larger than the laser light wavelength (10.6 nm), matter-light interaction is better described in the framework of ray optics. In this so-called Mie or ray optics regime forces occur due to the change in momentum of the light. The total force on a particle is the difference between the momentum flux entering the object and that leaving the object. The particles are pushed by the reflection/absorption of light from its surface while radiation forces due to refraction can pull a transparent object. SiO_2 is not transparent for the light emitted by CO_2 lasers. Therefore, the light is subject of reflection and absorption at the powder particles surface. In the present setup the resulting force pushes the SiO_2 particles antiparallel to the surface normal at the initial stages of the laser treatment. We cannot expect to observe any effect by this force in the present experimental setup. The energy of the incoming laser light dissipates into heat at the powder's surface. After prolonged irradiation the amorphous SiO_2 starts to melt. Now the interaction of the laser light with the microscopic entities of the liquid SiO_2, which are predominantly SiO_4 tetrahedra,[13] comes into play. Induced SiO_4 dipoles likely feel a force along the field gradient established by the laser light in the surface near region of the silica melt. Due to the strong absorption of the laser light in the silica, the light is not able to penetrate deeply into the liquid. The attenuation length of the CO_2 laser light in silica is approximately equal to its wavelength. In this scenario a field gradient is established not only parallel to the surface but perpendicular also. It can be assumed that the field gradient force drags molecular dipoles toward zones of highest electric-field strength, that is, highest light intensity. In the case of a pool of molten silica, a surface near migration of matter would not necessarily result in a significant net mass

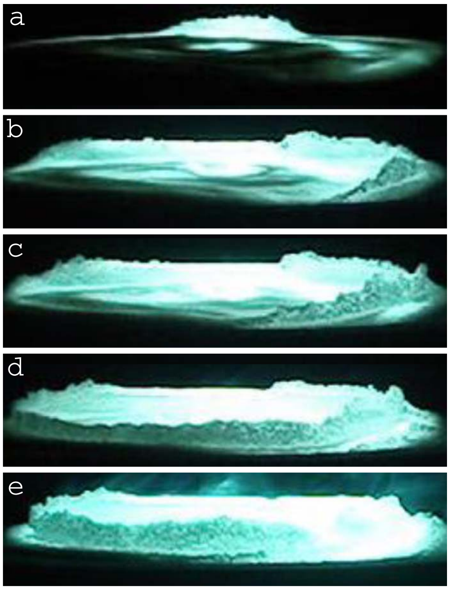

FIG. 2. (Color online) Sequence of photos taken as a function of the laser annealing time of a SiO_2 powder sample. The duration of the treatment is (a) 11 s, (b) 22 s, (c) 31 s, (d) 50 s, and (e) 67 s, respectively. For (a)–(d) the applied laser power output is 6 kW. For (e) the laser power output has been increased to 9.6 kW. It is noteworthy that significant SiO_2 evaporation is observed in (e) but not at lower laser power output in (a)–(d).

transport: equilibration between forces applied by the laser light and gravitational force and surface tension counteracts a significant net mass transport. Such a model could explain why areas with high laser intensity appear slightly elevated after an excessive treatment of a silica powder compact.

Bearing in mind that in our experiment surface protrusions are formed only on loose powder samples, but not on powder compacts, suggests that the surface tension is of general importance in the discussed scenario. The surface of a powder sample provides a negligible surface tension. Forces between the individual grains are negligible. In a sequence of pictures collected during the laser treatment of a powder sample, it is clearly noticeable that a sheet of partially molten powder peels off the surface in a way of curling and finally ends up almost parallel to the surface normal, see also Fig. 2. This migration of matter starts at points of highest laser intensity and then follows the circular shaped high intensity region of the laser spot on the sample. At the very beginning of the laser treatment it is difficult to judge whether the partially molten powder layers peeling off originates from lateral stresses induced by an inhomogeneous sintering of the powder or forces applied by the laser light. However, the fact that finally 7 mm high surface protrusions oriented almost parallel to the surface normal are formed, which even survive in the liquid state at excessive laser treatment, clearly argues against the suggestion that the protruding structures are formed by potentially based mechanical stress only.

In the present experiment, light induced forces become noticeable only when a SiO_2 melt exists at the powder samples surface. At a state where the laser treated powder sample combines minimum surface tension with just sufficient mobility due to the onset of SiO_2 melting we observe the formation of surface protrusions. Then, a silica melt particle mix is dragged toward high intensity areas on the samples surface. In this context, laser annealing experiments of SiO_2 powder compacts support the viewpoint that a low surface tension is a basic requirement for the formation of surface protrusions: no surface protrusions are formed in the initial stages of the laser treatment of powder compacts. In the case of a powder compact, the formation of surface protrusions in the initial stages of the laser treatment is inhibited by the existing van der Waals forces between the powder particles. When extended to the two dimensions of a surface, these forces can be interpreted as the surface tension of the powder compact also. Once a liquid surface is established at prolonged laser treatment of a powder compact, the formation of surface protrusions is prevented by the surface tension of the liquid SiO_2. On the other hand, forces applied by laser light are noticeable in a deep melt pool also as high laser intensity areas appear elevated relative to low intensity areas.

In summary we suggest a model in which light induced forces couple to molten silica. In this context, the formation of surface protrusions against gravitational force is enabled by the low surface tension of SiO_2 powder samples at the initial stages of the laser treatment where partially molten silica particles coexist with their melt. Once the protrusions are established they even survive the formation of a pool of liquid silica after an excessive laser treatment and, thus, withstand the SiO_2 melt surface tension. Understanding this self-sustaining effect would certainly provide more insights in the mechanisms governing the observed phenomenon. Further work will concentrate on this subject.

The question might arise as to why such a large area of silica has to be exposed to laser light in order to observe the described phenomenon. In fact, it should be possible to observe forces applied by laser light by annealing much smaller spots on a silica powder sample. Such experiments have been performed in the framework of the present study. In slightly smaller laser spots same results are obtained. However, at significantly smaller spot diameters silica droplets are likely formed as a result of the surface tension of the silica melt. From a droplet, however, it is difficult to judge whether its shape results from equilibrating gravitational force and surface tension or if additional light induced forces are involved.

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