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Bonding of glass with femtosecond laser pulses at high repetition rates

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Abstract We report on the welding of fused silica with ultrashort laser pulses at high repetition rates. Femtosecond laser pulses were focused at the interface of two optically contacted fused silica samples. Due to the nonlinear absorption in the focal volume and heat accumulation of successive pulses, the laser acts as a localized heat source at the focus position. Here, we analyze the influence of the laser and processing parameters on the amount of molten material. Moreover, we determine the achievable breaking stress by a three point bending test. With optimized parameters up to 75% of the breaking stress of the bulk material have been obtained.

1 Introduction

Fused silica has excellent mechanical, electrical, chemical and especially optical properties. Therefore, it plays a significant role for numerous applications. While the processing of fused silica or glass in general is relatively easy, the bonding to other materials is still a demanding application. Even the bonding of two glass samples is challenging, although numerous techniques have been developed. They are mainly based on well-known wafer bonding techniques [1, 2]. Some bonding methods use binders or glue as adhesives which, however, may lead to gas emission and aging of the bonds [3–5]. The most common bonding technique for glass is optical contacting. This is an adhesive-free process at which two ultra flat and ultraclean surfaces are brought together. The molecular attraction between the surface atoms adheres the samples. The bond strength is typically in the range of a few kPa and hence very weak. By thermal annealing the strength of these bonds can be increased due to the formation of strong covalent bonds [6, 7]. However, as the whole sample is heated tensions within the samples are induced This is especially severe if different glasses are bonded due to the difference of their thermal expansion coefficients. Another technique is to melt the interface with the help of laser radiation. In order to obtain absorption directly at the interface an absorption layer between the samples is required, which reduces the transparency [8].

Alternatively, ultrashort laser pulses can be used to induce local modifications inside transparent material due to non linear absorption [9, 10]. Therefore, the laser can act as a local heat source in the volume. Using high repetition rate laser systems operating in the MHz range leads to the melting of the focal region as well as a well-defined vicinity [11] due to heat accumulation of successive pulses [12–15]. The melting and solidification of the material results in the generation of covalent bonds if the laser focus is located at the interface between two adjacent samples [16, 17].

Figure 1 shows the laser induced modification at the interface of two circular blanks of fused silica. The laser induced circular modification area can be seen. Although the transparency of the modified area is reduced due to the induced material changes, it is easily possible to obtain high transmission in desired regions by welding only a certain area while leaving the rest of the sample unmodified. Additionally, by moving the laser focus along specific routes on the interface, tailored bonding adapted to custom designs becomes easily possible. Since no annealing step is neces-

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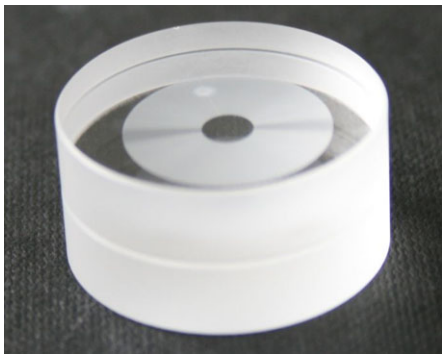

Fig. 1 Top view of two bonded circular blanks of fused silica. The *white lines* at the interface are the laser induced welds

sary to strengthen the bonds, even different materials can be joined. In addition the heat affected zone has only a height of a few µm. Thus the induced thermal stress in the sample is minimized. Due to the melting of the interface the welds offer the possibility of gas proof sealings. In this paper we investigate the achievable bond strength when welding fused silica substrates with ultrashort laser pulses at high repetition rates. For a quantitative determination of the bond strength we apply a three point bending test.

2 Experimental setup

For the laser welding we used a femtosecond oscillator providing pulses at a wavelength of 1030 nm, a repetition rate of 9.4 MHz, an average output power of 5 W and a pulse duration of 550 fs (Amplitude Systems, t-Pulse 500). The repetition rate and pulse energy were varied by an external acousto-optic modulator and a halfwave plate followed by a polarizer, respectively. For the experiments, we used the second harmonic (515 nm) generated in an LBO crystal. Commercially available fused silica circular blanks (Layertec) with a diameter of one inch and different thicknesses were used as glass samples. As the height of the molten and ejected material is only in the range of a few hundred nanometer [18] the samples were optically contacted before the laser welding process. This requires a sample roughness of 2 nm and a flatness below 125 nm. The laser pulses are focused with aspheric lenses with numerical apertures of 0.1 up to 0.4 depending on the sample thickness. In order to position the focal point at the interface, we measured and minimized the size of the reflected light spot on the sample surface. Afterwards, we moved the lens downward according to the optical thickness of the upper sample. With the help of a high-precision three axis positioning system (Aerotech, ALS 130-150) both, the sample and the focusing optic, could be moved to realize different writing geometries within the sample. The complete setup is sketched in Fig. 2.

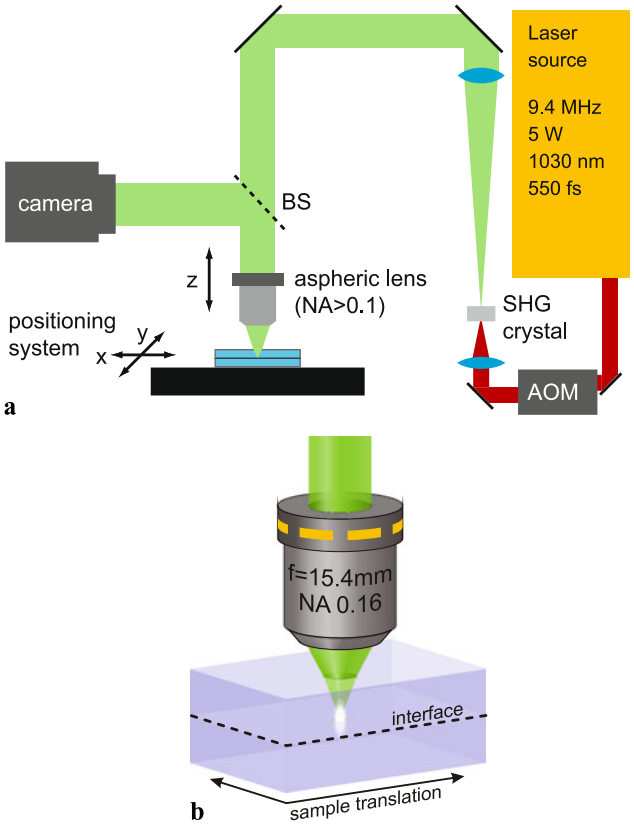

Fig. 2 (**a**) Experimental setup for the laser welding process. The laser pulses of the oscillator are converted to a wavelength of 515 nm. To control the repetition rate an external acousto-optic modulator (AOM) was utilized. The beamsplitter (BS) and camera were used to control the focus position on the sample. (**b**) Schematic of the inscription process. The laser pulses are focused at the interface between two glass samples by an aspheric lens. The sample is translated with respect to the laser focus by a high-precision positioning system to realize different bonding geometries

For the determination of the breaking stress of bonded samples typically a blade test is used [19]. Here, a blade is inserted at the interface and the size of the disjoined area is measured. However, this test is unsuitable for only partially bonded samples. Furthermore, even small variations of the measured size lead to high fluctuations of the breaking stress results. Therefore, we used a three point bending test to measure the force F necessary to break the bonded samples and hence calculated the breaking stress. After the welding process, we cut the blanks into rectangular rods. In a next step, the bonded interface is placed at the center between two supports directly below the stress pin applying a defined force F. According to [20], the breaking stress σ is calculated by

$$\sigma = \frac{3 \cdot F \cdot l}{2 \cdot b \cdot h^2},$$

where b, h and l refer to the setup geometry as sketched in Fig. 3 (in our case: $b = h = 4$ mm, support span $l = 10$ mm).

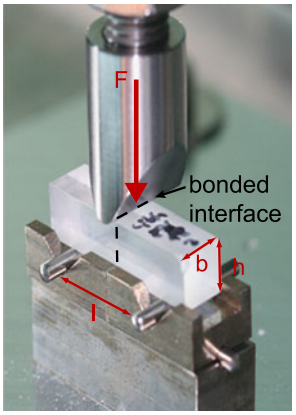

Fig. 3 Three point bending test setup to measure the bond stability. Under the stress pin one can see the rectangular glass sample, which was cut after the welding process

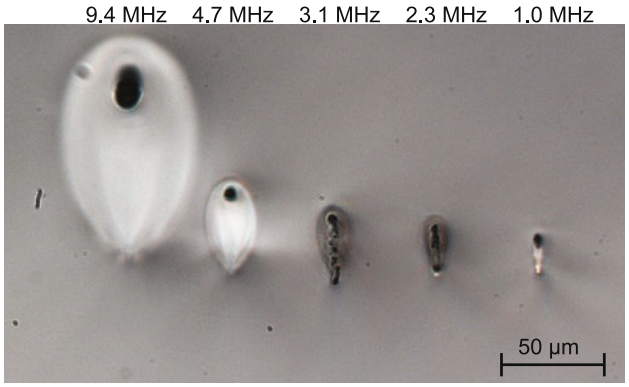

Fig. 4 Side view on the laser induced modifications in fused silica for different repetition rates at a pulse energy of 120 nJ (NA 0.55) and a translation velocity of 20 mm/min. At lower repetition rates the modification zone is smaller due to the reduced heat accumulation. The pulse absorption starts at the bottom of the modification. The *black spots* within the molten zones are voids, resulting from microexplosions

For these measurements, the most important sources of error during these measurements are based on defects and scratches at the surface of the rods induced during the cutting process. Also defects within the sample play a significant role for the crack propagation and the force required to break the sample. Thus, the estimated error of breaking stress measurement for bulk material is about 25%. However, for laser welded samples the original interface is a predetermined breaking plane, which lowers the error of the measurement. Multiple measurements indicate an error of about 15%.

3 Results

The size and shape of the molten region depend on the laser and material parameters as well as the sample translation velocity and changes in the absorption caused by the generation of free electrons. Due to aberrations, the focusing depth also affects the shape of the molten volume [21]. A cross section for a typical thermally induced modification shape is shown in Fig. 4 at different laser repetition rates. Here, the modifications were placed in the bulk material at a depth at which the aberrations of the lens are minimal. The absorption starts at the lowest point of the bulb shape indicating the location of the focal point. At the beginning initial seed electrons are produced by nonlinear ionization. Since these free electrons can linearly absorb single photons the position of main absorption moves toward the focusing lens and the laser modification volume becomes elliptic [11]. We observed an absorption threshold of about 10 J/cm^2 for a pulse duration of 550 fs and 515 nm wavelength (NA 0.25, 135 nJ). This threshold is independent of the translation velocity from 1 to 200 mm/min. Due to the thermal expansion of the heated focal region and the fixed outer boundaries, high pressure occurs resulting in microexplosions and void-formation (black regions within the molten area in Fig. 4) [22, 23].

In the outer region of the laser modifications one can see the effect of the heat accumulation, where material outside the laser focus was heated above the melting point. Therefore, the molten zone is much larger than the focal volume in this case. After laser irradiation the molten material cools rapidly, leading to a non-uniform resolidification [24]. Note that the sample in Fig. 4 was translated at 20 mm/min, leading to several thousand laser pulses incident per µm. Figure 4 shows decreasing molten volume with decreasing repetition rate due to the lower heat accumulation. Because of the large heat conductivity of fused silica, no melt occurs in the material surrounding the focal volume at or below a repetition rate of 1 MHz. However, voids can be formed due to the high peak power in the focus. The exact process of melting and void-formation is very complex and still not fully understood.

For laser welding the laser focus was positioned at the interface between two optically contacted samples. After welding we broke the samples apart and measured the size of the molten zone for different laser parameters. In Fig. 5 one can see a dark field microscope image of the ripped interface. Here, the bright spots originate from the residues of the welded material. The lines were written with a pulse energy of 160 nJ, a repetition rate of 9.4 MHz and different translation velocities.

The areal density of the molten material at the interface increases with higher translation velocity, as can be seen in Fig. 5. Although the energy deposited per spot decreases with higher translation velocity the width of the molten lines is nearly constant. The welding seams are disconnected due to the formation of voids during the laser welding process. Therefore, we do not only consider the width of the lines but an effective molten area. To quantify the amount of molten material, we wrote welding seams with different laser parameters. Figure 6 shows the influence of the laser repetition rate and translation velocity on the amount of molten

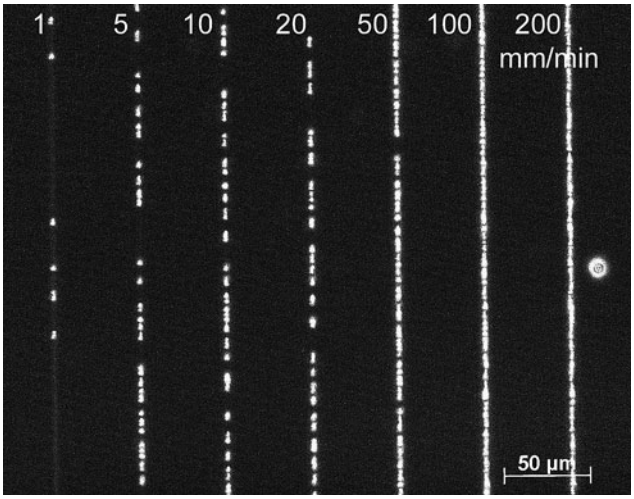

Fig. 5 Dark field microscope image of the welding seams in fused silica after breaking the samples apart. The bright regions, indicating residues of the welded material, increase with the translation velocity of the sample. The lines were written with a pulse energy of 160 nJ, a repetition rate of 9.4 MHz and different translation velocities

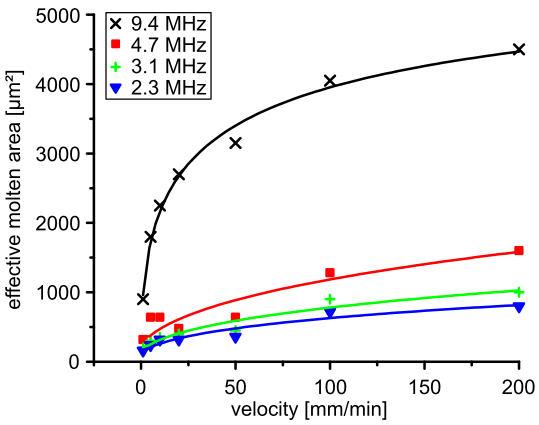

Fig. 6 Dependence of the effective molten area at the interface of laser bonded fused silica on the translation velocity and the laser repetition rate for an energy density of 35 J/cm^2. As the molten lines are disconnected (see Fig. 5) we considered a molten line for a length of 1 mm and measured the amount of molten material

material for a pulse energy of 100 nJ and an energy density of 35 J/cm^2 (NA 0.55). In all cases, we measured an increased effective molten area at higher translation velocity. This may be explained with a reduced formation of voids at the interface of the samples. At slow velocities very high lattice temperatures are reached resulting in microexplosions in the focal region and void-formation. These voids present vacancies of the molten material. Furthermore, the induced voids scatter the subsequent laser pulses and less energy is deposited in the focal volume leading to less melting. Therefore, the molten area at the interface is reduced due to the void-formation. At higher translation velocities the maximum temperature is lower, the void-formation is reduced and more material at the interface is molten.

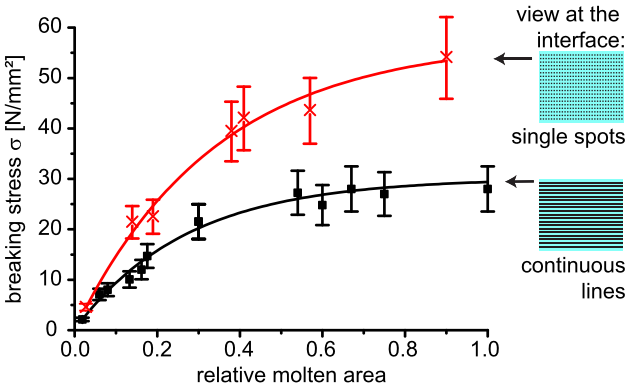

Fig. 7 Dependence of the breaking stress on the size of the molten area. Here, the molten area was normalized to the interface. The breaking stress saturates at different values depending on the processing geometry

In addition, we measured an increasing amount of molten material with increasing repetition rate. This is due to stronger heat accumulation, when the deposited power increases, as seen in Fig. 4. Nevertheless, the increase is strongly nonlinear: the molten area increases by a factor of three, while the repetition rate is only doubled from 4.7 to 9.4 MHz.

In the next step we measured the breaking stress for different molten areas at the interface with the three point bending test. The measured breaking stress increases with increasing size of the molten area, as shown in Fig. 7. When the individual welds consist of continuous molten lines the initial linear increase of the breaking stress finally saturates to a maximal value of 25 N/mm^2. This is about 40% of the breaking stress of bulk fused silica (70 N/mm^2). To understand this, one has to consider the crack formation within the breaking plane and the surface energy which is necessary for the crack propagation. On the one hand, the molten lines establish strong bonds between the glass samples but on the other hand, they act as predetermined breaking points. If these lines are continuous only a small amount of surface energy has to be provided for an initial crack to propagate along the whole line. To prove this hypothesis we used single spots as welding points. In Fig. 7 one can see that these single spots offer a much higher breaking stress than continuous lines. This is caused by the maximized amount of borders. Each of these borders form a resistance to the crack propagation and thus a higher surface energy to crack the whole interface is required. If the density of these discontinuous lines is optimized, a maximum breaking stress of 54 N/mm^2 may be reached, which is about 75% of bulk fused silica. This is in very good agreement with measurements in which we applied the same processing to the bulk material. In this case we measured a breaking stress of 58 N/mm^2.

To explain the fact that the breaking stress is limited to approx. 75% of the bulk material we took a closer look at

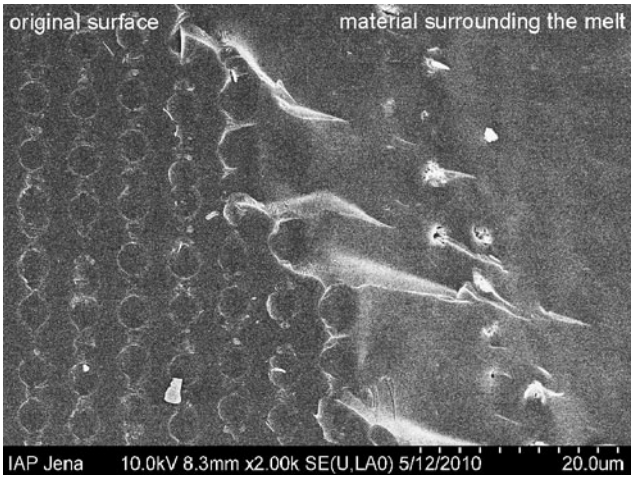

Fig. 8 SEM image of a broken interface of two laser bonded samples. Note the change in the crack position from the original surface into the bulk material

the ripped interface after the three point bending test. For a small molten area, where a linear dependence between the breaking stress and molten material is observed, the crack propagates directly at the original interface. This is proved by the remnant of the molten material which resides at the ripped interface, which one can see in Fig. 8 at the original surface. These segments may occur either as bulges or as shallow holes with a height of about 200 nm. As long as the distance between the molten lines is larger than 5 µm the crack propagation occurs mainly at the interface. Starting with a surface coverage of molten material greater than 60% the crack rather propagates in the surrounding material than along the original interface. The new breaking plane has a distance of several micrometers to the original interface. In Fig. 8 the change in the breaking plane from the interface into the material can be seen. These results can be attributed to the induced stress in the material during the welding process. Due to the melting and subsequent quenching, tensions are induced in the modified zone as well as in the surrounding material. During the solidification not all of these stresses can relax. These tensions act as predetermined breaking points. If the distance between the molten zones is small enough for the induced tension fields to overlap, a complete predetermined breaking plane is formed. This explains the limitation of the achievable breaking stress. Due to the formation of new strong bonds at the interface between the samples, the interface is no longer the preferred plane for the crack propagation. Instead of this, a plane along the tension fields acts now as breaking plane.

4 Conclusion

Ultrashort laser pulses at high repetition rates offer the possibility to weld glasses and form strong bonds. In fused silica we could obtain a breaking stress up to 75% of the bulk material by optimizing processing conditions. This is in good agreement with the breaking stress for similar modifications induced in the bulk material. Such strong bonds offer the possibility for localized and application specific bonding of glasses.

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