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Single femtosecond laser pulse-single crystal formation of glycine at the solution surface

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ABSTRACT

We demonstrate femtosecond laser-induced crystallization of glycine from its supersaturated solution depending on laser tunable parameters (pulse energy and repetition rate) and focal position, and examine the crystallization probability, crystal morphology, and crystal polymorph. The generation of cavitation bubble through multiphoton absorption of water depends on input laser pulse energy and repetition rate, which strongly determine morphology and number of the obtained crystals. Significant increase in the crystallization probability is observed by irradiating the femtosecond laser pulses to the air/solution interface, and single pulse-induced single crystal formation is successfully achieved. The crystallization mechanism is discussed in view of inhomogeneous mechanical stress induced by cavitation bubble generation and molecular assembly characteristics of the surface.

1. Introduction

Light-induced crystallization in supersaturated solutions of organic and biological molecules has been a very attractive topic in the fields of crystal science and engineering [1–16]. In particular, utilizing continuous wave (CW) or nanosecond pulsed laser has been reported on relatively small molecules. For a representative example, Garetz and co-workers demonstrated the crystallization of organic molecules, notably amino acids, with 1064- or 532-nm nanosecond pulse lasers and explained that the nucleation proceeds due to optical Kerr alignment of the molecules based on an intense electric field of the laser light [2,3]. They also succeeded in controlling crystal polymorph by switching polarization of laser light. On the other hand, our group demonstrated laser trapping-induced crystallization of glycine by focusing a laser beam at air/solution interface of the solution [8,9]. Tightly focused CW near-infrared laser beam was employed as a trapping light source to gather molecules or clusters into the focal volume, where the high concentration area was consequently formed, eventually leading to the nucleation. Note that this laser-trapping crystallization is achieved only by focusing the laser beam at the air/solution interface, never in solution. Thus, we consider that this focal position is very critical for laser-induced crystallization. In view of spatial and temporal control of crystallization, laser irradiation is one of the most promising approaches and it is expected that the change in the irradiation position and the adjustment of laser parameters such as wavelength, intensity, repetition rate, polarization can give an optimal condition to prepare fine crystals with high quality and a desired morphology.

Besides the laser-induced crystallizations mentioned above, we have also reported that the irradiation of an intense near-infrared femtosecond laser pulse can trigger molecular crystallization [12–15]. When the intense femtosecond laser is focused into their solutions, morphological phenomena such as shockwaves, cavitation bubbles, and jet flow, and the sequential photomechanical ablation are induced due to multiphoton absorption of solutions. Previous studies suggest that the bubble formation and expansion cause transient pressure in the vicinity of the focus, triggering the nucleation. This crystallization mechanism is completely different from other laser-induced crystallization because only this technique triggers nucleation by laser irradiation to solvent.

Here, we investigate on how femtosecond laser parameters and the irradiation position control crystallization probability and

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morphology by examination of glycine crystallization in the supersaturated aqueous solution. Glycine is a typical molecule for studies on molecular crystallization mechanism, so that several groups studying the laser-induced crystallization have employed it as a standard sample [2,3,8,9]. We have examined the repetition rate, pulse energy, and focal position dependences, and show that the frequency of cavitation bubble generation strongly affects crystal morphology. The further investigation on glycine crystallization depending on laser parameters such as the power and shot number will be helpful to optimize them in order to obtain fine crystals for X-ray crystallography, and eventually we have succeeded in demonstrating single laser pulse-single crystal formation.

2. Experimental section

Glycine (99.0%, Wako Chemical) was used without further purification. We prepared 0.5 ml of glycine aqueous supersaturated solution in 1.5 ml glass bottle with 4.7 mm of bottom diameter (Nichiden Rika Glass). The solution was adjusted with concentration ranging from 3.0 to 4.0 M, which corresponds to supersaturation degree (SS) ranging from 1.0 to 1.33. Here, SS was defined as C/C_{sat}, in which C and C_{sat} (3.0 M at room temperature of 22 °C) are actual solute concentration and the saturated solute concentration at a given temperature, respectively [17]. The mixtures of glycine and water were heated up to 60 °C in water bath for complete dissolution of solutes, and then were slowly cooled down to room temperature. The solution thickness in the bottle was measured to be approximately 3.0 mm.

After confirming the absence of spontaneous crystallization from 5 to 8 days, the sample was put on an inverted microscope (IX-71, Olympus) sample stage and was exposed to linearly polarized femtosecond laser pulses (wavelength; 800 nm, pulse duration; 160 fs, Spitfire Pro, Spectra Physics) through an objective lens (10×, N.A. 0.25, PlanN, Olympus) as shown in Fig. 1. Pulse energy was adjusted by using a half-wave plate, a polarizing beam splitter, and a variable neutral density filter, and was measured throughout the objective lens by an energy meter (842-PE, Spectra Physics). The repetition rate of femtosecond laser pulse train was controlled by a Pockels Cell. The crystal formation and crystal morphology were observed with a digital CCD camera (CV-S3200N, JAI) attached to the microscope. All the experiments were carried out at room temperature. The experiment was repeated for 10 samples under each experimental condition, and crystallization probability was estimated by counting the number of crystallization samples from them.

X-ray diffraction and crystallographic analysis were carried out to examine and characterize the prepared crystals. X-ray diffraction data were collected on a Bruker APEX DUO diffractometer equipped with the APEX-II 4K CCD detector and kappa 4-axis goniometer with graphite monochromated Mo-Kα radiation (λ=0.71073 Å) at 100 K. Intensity data were corrected for Lorentz and polarization effects. Unit cell parameters and orientation matrix for the data collection were determined from least-square refinement of diffraction data. Data reduction was carried out using 'Bruker SAINT' program.

3. Results and discussion

3.1. Crystal morphology depending on repetition rate

Fig. 2 shows how crystallization probability and crystal morphology depend on the repetition rate of irradiated femtosecond laser pulse train. Pulse energy, and irradiation time were fixed to 100 μJ/pulse (19 PW/cm^2 per pulse) and 10 min, respectively. In this case, we focused the laser pulses at several microns above the glass surface to avoid laser ablation of the glass. After a certain time from the femtosecond laser pulse irradiation into glycine supersaturated solution, we successfully obtained glycine crystals and found them in the glass bottom. It was found that the crystallization probability increased with the repetition rate, and that it became saturated to 100% above 250 Hz. On the other hand, at the low repetition rate less than 5 Hz, no crystallization took place. This implies that the crystallization probability is strongly related to the number of irradiated pulses, and it should be lower than 100% per pulse.

In addition, we examined whether or not crystal morphology exhibited strong correlation with the repetition rate (Fig. 2(b) and (c)). After irradiation at lower repetition rates, the transparent single crystals with prismatic morphology trapped at the air/solution interface were obtained frequently. We suggest that the single crystal was floated by the generated cavitation bubble up to the solution surface. The crystal size (approximately 5 × 4 × 3 mm) and morphology are almost reproducible under the same experimental conditions, so that the size is probably determined by concentration of glycine, suggesting an important role of spontaneous crystal growth after nucleation. On the other hand, we found powder-like crystals, i.e., small, uniformly sized (mean size 50 × 20 × 10 μm), and a number of the crystals at higher repetition rates. At the middle rate around 50 Hz, about 5–10 polycrystals with about 2 × 1 × 1 mm were dominantly observed. Also we found that the crystal morphology is dependent on the necessary time for crystal formation. With decreasing the size of crystal, the formation time becomes shorter. The single crystal formation took about 2–3 days, whereas the powder-like crystals could be formed during pulse irradiation, that is, within 10 min.

Such repetition rate dependence of crystal morphology has been previously reported on lysozyme crystallization by our group, sometimes accompanied with the denaturation depending

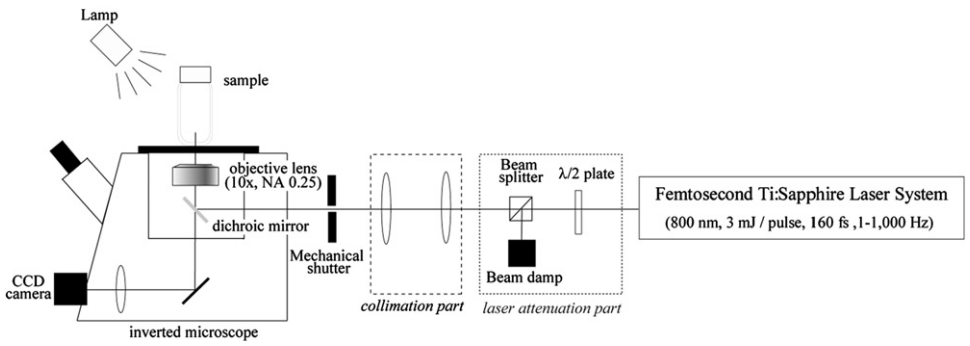

Fig. 1. A schematic illustration of experimental setup for femtosecond laser-induced crystallization.

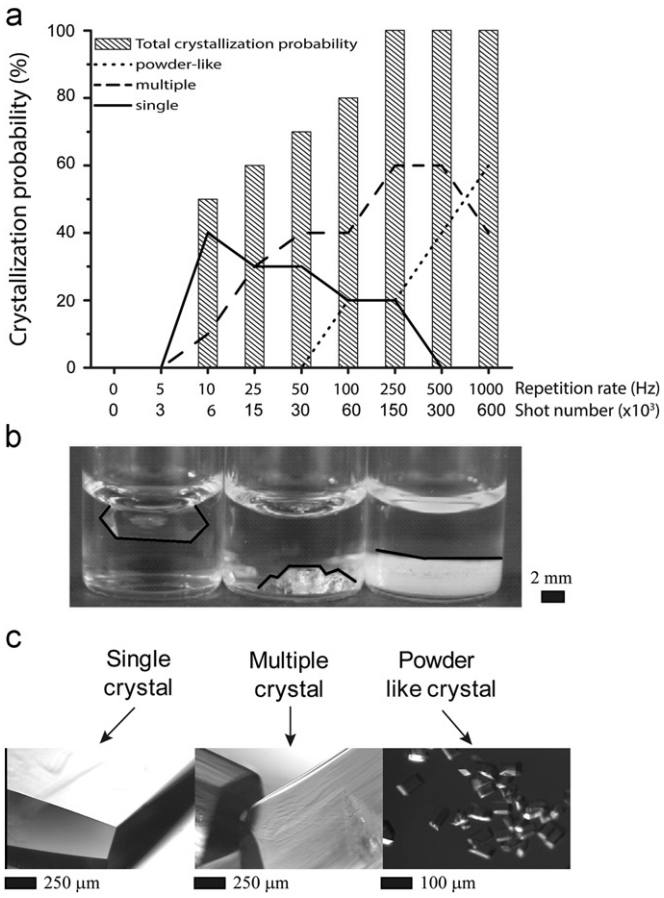

Fig. 2. (a) Femtosecond laser repetition rate dependence of crystallization probability of glycine. For each condition, we irradiated the laser pulses to 10 sample solutions. Pulse energy and irradiation time were fixed to 100 μJ/pulse (19 PW/cm^2 per pulse) and 10 min, respectively. The concentration of glycine was 4.0 M (SS 1.3). Dotted, dashed, and solid lines plotted in the graph correspond to each crystal morphology of powder-like, multi crystals, and single crystal, respectively. The definition of each crystal form is explained in the text. The formation probability of each crystal morphology is given as a ratio of the number of each crystal morphology to the total number of crystal-forming samples. (b) and (c) Photographs and optical microscope images showing typical crystal morphology which were obtained with the pulsed laser irradiation with different repetition rate irradiations in figure.

on the repetition rate [12]. For the powder-like crystal formation, we consider that the frequent nucleation caused by a high repetition rate can be regarded as homogeneous nucleation, resulting in the powder-like crystal formation. Here it should be noted that we could directly observe the crystallization during laser irradiation only in the case of powder-like crystal formation. It is suggested that further laser irradiation after the crystallization leads to laser ablation of the formed crystal, which crashed the formed crystal into large amount of the small fragments. The fragments can grow quite rapidly, resulting in providing the powder-like crystals. Namely, powder-like crystals can be observed in the focal volume before its floating up to the solution surface because of the rapid growth. Actually, when the femtosecond pulses were directly irradiated to the glycine crystal generated spontaneously in the bottle, the fragmentation of the crystal surely takes place through its laser ablation induced by multiphoton absorption [18]. In contrary, laser irradiation at lower repetition rates preferentially provided the single crystals. The growth rate of the single crystal is much slower than that of the powder-like one, so that the crystal can be identified only after being floated up to the solution surface.

Needless to say, single crystal formation is more acceptable for X-ray crystallography prior to powder-like crystal, so that laser irradiation with a lower repetition rate should be preferable. At the lower repetition rate, the probability of nucleation per unit time should be lower because the interval for the nucleation becomes longer, and the probability that the crystals stepping away from the focal point will increase. This may cause inhomogeneous distribution of crystals size and difference of surface free energy among the plural faces of the crystals, which lead to single crystal formation as the result of Ostwald ripening. This is consistent with longer time for formation of the single crystals as mentioned above. Consequently, the results of this repetition rate tell us how we can adjust laser parameters for obtaining single crystals.

Concerning crystal polymorph, we found that all the obtained crystals with different crystal morphologies, i.e., single, multiple, and powder-like crystals, could be assigned as α-form, which were confirmed by analysis of X-ray diffraction (see Fig. 1S in supplement data). It was also confirmed that the unit-cell parameters of all samples was fully consisted with those of α-form glycine crystal reported by Iitaka previously [19]. It is well known that only α-form, which is the kinetically most accessible phase, is obtained through conventional crystallization method of re-crystallization from H_2O [20]. Garetz and co-workers successfully demonstrated the γ-form crystal formation by linearly polarized nanosecond laser pulses irradiation into the solution only with the SS range more than 1.53, not with lower SS [21]. Considering the high laser intensity (PW order) of femtosecond laser used in this experiment, which is 6 orders higher than that of nanosecond laser, the γ-form crystal formation was possibly expected in this experiment, but only α-form crystal was confirmed under any experimental conditions.

3.2. Laser bubbling leading to crystallization

Fig. 3 shows pulse energy dependence of glycine crystallization probability. With increasing the pulse energy, crystallization probability increased, while it showed a threshold of crystallization around 2.5 μJ/pulse (0.48 PW/cm^2 per pulse), which was in good agreement with that of cavitation bubble generation observed by CCD camera observation. This implies that the laser-induced cavitation bubble triggers crystal formation. Recently,

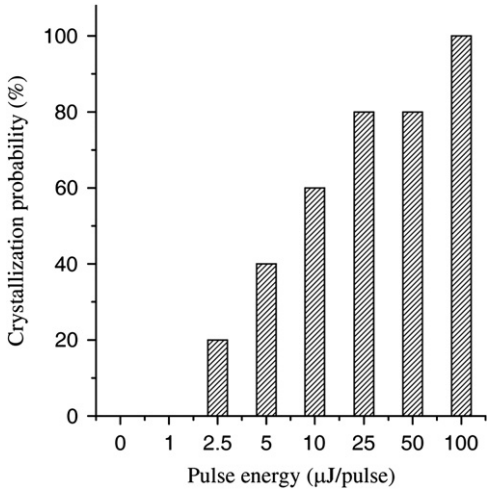

Fig. 3. Femtosecond laser pulse energy dependence of crystallization probability of glycine. The repetition rate, irradiation time, and the focal position were fixed to 100 Hz (pulses/second), 10 min, and at the solution/glass interface, respectively. The concentration of glycine was 4.0 M (SS 1.3). For each condition, we irradiated the laser pulses to 10 sample solutions.

high-speed camera observation of crystallization process upon laser-induced cavitation generation revealed that a film-like crystal of anthracene was created at the bubble surface [15]. Similarly, highly concentrated area of fluorescent protein [16] and optical disturbance of $KMnO_4$ [22] along with the laser-induced bubble surface were reported upon laser irradiation in their solutions. All these results support an idea that the expansion and collapse of the cavitation bubble can form local initiate high concentration area of the molecules around the bubble surface, leading to molecular nucleation. Therefore we conclude that the cavitation bubble generation triggers nucleation of glycine.

Mass spectrometry analysis for the obtained crystal as shown in Supplementary material possibly revealed that thermal and photoproducts generated upon laser irradiation was not generated during laser irradiation. Therefore, we consider that the crystallization takes place not through photochemical process [6,7], but through various femtosecond laser-induced phenomena such as cavitation bubble and shock wave. The femtosecond laser can excite not only water but also glycine via multiphoton absorption [18].

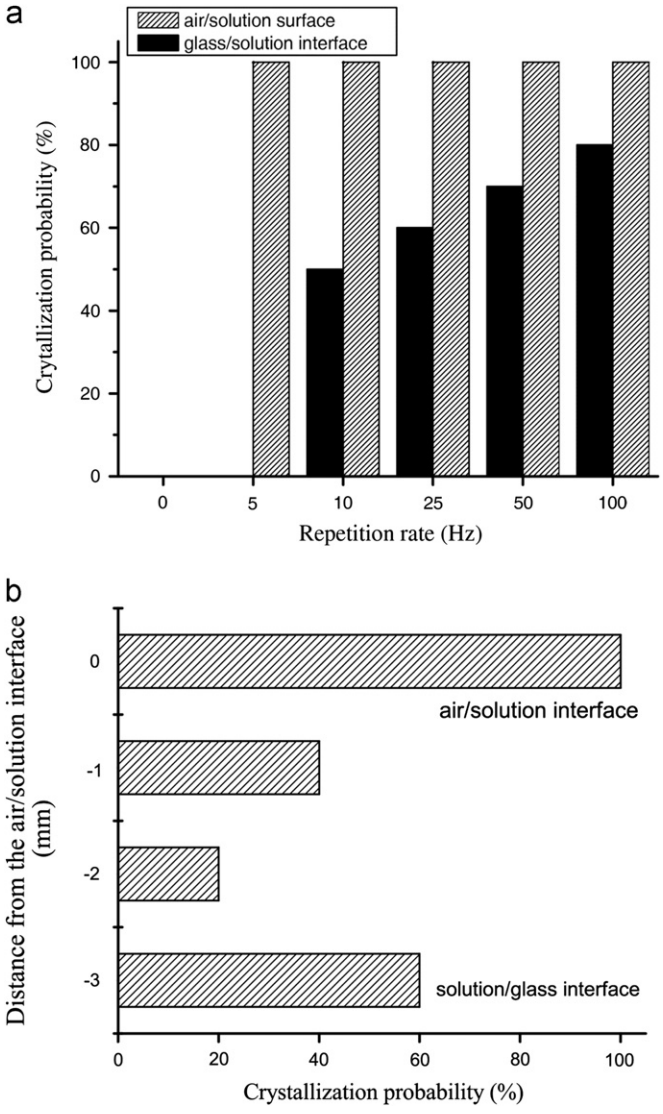

Fig. 4. Focal position dependence of crystallization probability of glycine. (a) Comparison between femtosecond laser irradiation at air/solution interface (grey) and at solution/glass interface (black) with different repetition rates. Crystallization probability at solution/glass interface irradiation is taken from Fig. 3 for reference. (b) Crystallization probability at different focal positions. The air/solution interface was regarded as 0 μm height. For each condition, pulse energy was 100 μJ/pulse (19 PW/cm^2 per pulse) and irradiation time was 10 min. The concentration of glycine was 4.0 M and we examined 10 sample solutions for each condition.

3.3. Efficient crystallization at the solution surface

Next, we elucidated focal position dependence of the crystallization probability and summarized the results in Fig. 4. It was found that crystallization probability was improved upon laser irradiation at air/solution interface (Fig. 4(a)). Note that huge numbers of powder-like crystals are formed even in the lowest repetition rate (5 Hz), which is in contrast to the result on the above repetition rate dependence upon conventional irradiation inside the solution. This means that we can reduce laser exposure time, which may hopefully result in single crystal formation. In order to confirm the interface effect, we shifted the focal position from the air/solution to the solution/glass interface by 1 mm step (Fig. 4(b)). As the result, it was found that the crystallization probability was increased by irradiation into both interfaces, and that the probability was the highest at air/solution interface. In contrast to their interface irradiation, the irradiation inside the solution suppressed the probability. Therefore, we conclude that the irradiation at air/solution interface is more efficient on femtosecond laser-induced crystallization.

The present results are possibly explained from two viewpoints. Firstly, we suppose that glycine molecules are adsorbed at the interfaces and they may be better aligned for leading to decrease in free energy barrier between solution and crystal phase [9,23,24]. Although glycine shows negative adsorption on water surface, glycine molecules may contact to the interface frequently in the supersaturated solution and form ordered domains, where molecules are aligned easily undergoing the nucleation. Secondly, cavitation bubble accompanied with transient mechanical stress should not be symmetrical and spherical at the air/solution and solution/glass interfaces, [25,26] although it expands symmetrically inside the solution. Especially, at the air/solution interface, a unique solution flow is created from the surface because it can be regarded as free boundary. Thus, the surface deformation corresponds to bubble formation inside the solution, which may enhance crystallization probability, although no approvable heating effect in our femtosecond case. We consider that the molecular alignment characteristics of solution surface and the surface deformation are coupled with each other, leading to the more efficient crystallization at air/solution interface.

Because the improvement of crystallization probability via femtosecond laser irradiation at interfaces was found, it can be expected to obtain crystals from lower supersaturation concentration. Fig. 5 shows crystallization probability depending on SS at different focal positions, where pulse energy and irradiation time were fixed to 50 μJ/pulse (9.5 PW/cm^2 per pulse) and 10 min, respectively. At lower SS less than 1.2, crystal formation was found only in the case of the surface irradiation. On the contrary, at higher SS around 1.33, which corresponds to 4.0 M, the crystal appeared on both irradiation at the surface and inside the solution (1 mm above the bottom), although the probability was much higher in the case of the surface irradiation. This result is consistent with the result of repetition rate dependence shown in Fig. 3.

3.4. Individual formation of single crystal

Finally, the laser exposure time dependence, i.e., pulse number dependence of crystal morphology was examined. The femtosecond laser pulses were focused at the air/solution interface of 4.0 M with different exposure times of 10, 30, and 180 s at 5 Hz,

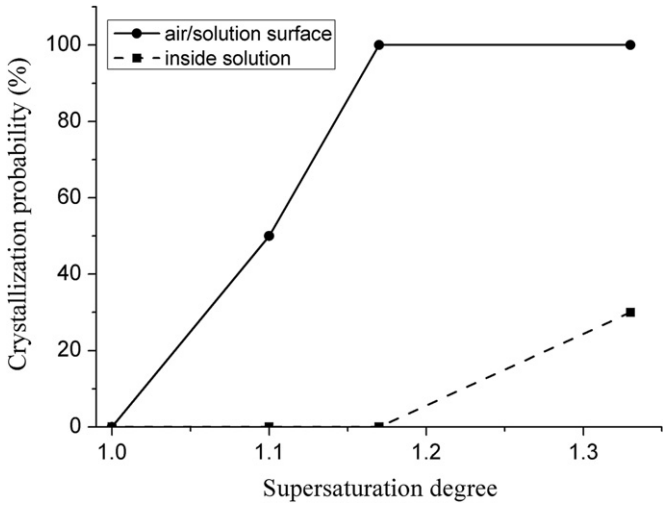

Fig. 5. Glycine concentration dependence of crystallization probability at different focal positions; the air/solution interface and inside the solution (1 mm above the bottom). The concentration of glycine was converted to SS by dividing the concentration by the saturated one (C_{sat}=3.0 M). For each condition, pulse energy was 50 μJ/pulse (9.5 PW/cm^2 per pulse) and irradiation time was 10 min, and we examined 10 sample solutions for each condition.

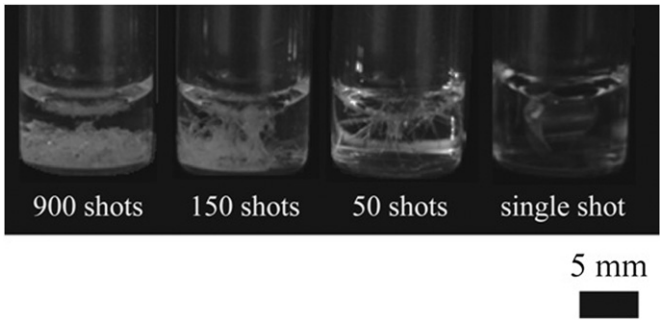

Fig. 6. Photographs of crystal morphology obtained with different numbers of pulse irradiation at the air/solution interface. The pulse number was tuned as 900, 300, 50, and single shot, respectively. The concentration of glycine was 4.0 M and pulse energy was adjusted to 100 μJ/pulse (19 PW/cm^2 per pulse).

corresponding to 50, 150, and 900 shots irradiation, respectively. Single shot irradiation was also carried out. It is important to note that the low pulse repetition rate employed here could not induce crystallization in the case of irradiation inside the solution. Fig. 6 represents the crystal morphologies depending on the shot number upon irradiation at the surface. Crystals were clearly formed in each case and it is obvious that that the number of crystals and their morphologies are strongly correlated with the pulse number. For larger pulse number, 900 shots, multi crystals were found as similar to the result shown in Fig. 2. With decreasing the pulse number, the shape of crystal became longer and slighter, while the number of crystals decreased. At 50 shots irradiation, needle like crystals, whose mean size is around $4 \times 1 \times 1$ mm, were found, this is characteristic of this lower pulse number irradiation. Eventually for single shot irradiation, it was found that a single crystal was formed and floating on the solution surface at a certain probability (about 10%). It is important that the single pulse irradiation forms only one single crystal. All the crystals except the single crystal were formed within several minutes after laser irradiation, while the single crystal formation took about 2–3 days.

It is difficult to decrease the total pulse number because the lower shot number provides lower crystallization probability. However, the air/solution interface can make the crystallization probability high, so that single laser pulse irradiation-single crystal formation is realized as demonstrated here. Here it should be emphasized that the single pulse-single crystal formation was not made possible in the case of laser irradiation inside the solution. Consequently, by examining the laser parameter effect on glycine crystallization, we have for the first time succeeded in single crystal formation by single femtosecond shot irradiation especially by utilizing the surface irradiation.

4. Conclusion

In summary, we have demonstrated femtosecond laser-induced crystallization of glycine in its supersaturated H_2O solution. We found that morphology of crystals prepared by cavitation bubble formation upon the femtosecond irradiation into solution was determined by laser repetition rate. Especially, lower repetition rate of femtosecond laser irradiation tends to result in single crystal formation. We also found increase in crystallization probability at air/solution interface, which means an important interplay between molecular adsorption and spatial limitation of mechanical stress induced by cavitation bubble formation. Such findings help us to identify crystallization conditions for other compounds. Hence, as the results of optimizing laser parameters based on our investigation, we finally can find a condition under which single crystal formation with high probability (10%) is made possible by single shot irradiation to the air/solution interface. Here we can propose that optimizing laser parameters, especially utilizing the solution surface will give more sophisticated crystallization technique. In particular, protein crystallization at the air/solution interface is highly expected because the high crystallization probability through conformation change of protein can be simultaneously induced at the solution surface [27]. To confirm the effect of surface irradiation more directly, we will use high speed camera measurement to observe the behavior of cavitation bubble in/on glycine solution and employ surface selective spectroscopy such as sum frequency generation (SFG) to confirm glycine structure during adsorption.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.jcrysgro.2012.11.018.

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