



# Investigating Crystal Nucleation and Growth under the influence of Optical Tweezers

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Optical tweezers allow crystallisation from solution to be localised to a known position within a relatively well-known timeframe [1]. However, the impact optical tweezers can have on crystallisation still requires investigation. Depending upon the makeup of the solution under investigation, this can result in significant localised heating surrounding the tweezing focal volume [2]. One commonly used way to minimise this temperature rise in aqueous solutions is to use deuterium oxide. However, since this is not the standard isotopologue of water, the fundamental impacts that this would have on bulk crystallisation have not been previously detailed beyond solubility. Investigations show a reduction in the primary nucleation rate. Crystal growth from supersaturated solution can be enhanced locally through the presence of an optical tweezing focus even a relatively low powers. However, the localised growth of a macroscopic crystal can also be inhibited by using regular water and a laser wavelength known to be highly absorbing at a high enough power. The presence of a trapped silica particle appears to prevent the tweezers from being able to increase the growth of a crystal close to the focus in conditions where growth would be enhanced in the absence of the silica particle. At higher laser powers, the focus can directly induce nucleation from solution even in undersaturated conditions when near a solution-air interface. But when moving into the bulk of the solution, the ability to nucleate from the solution diminishes with increasing distance between the interface and the droplet edge. Future work will explore the impact that commonly used tweezing particles have on primary nucleation characteristics, and the effect that holding particles within the focus of the tweezers have on their ability to influence nucleation directly.

## Tweezer Induced Heating

Previous experiments have shown that the focus of an optical tweezer can cause the region surrounding the focus to experience temperature elevation. However, this is dependent upon several factors, the main one being absorption. One way to significantly reduce heating in aqueous solutions is to use deuterium oxide (D<sub>2</sub>O) due to the orders of magnitude reduction of absorption coefficient between the isotopologues at common trapping laser wavelengths [2]. Modelled for the maximum possible temperature rise at 60 microns from the heat sink.

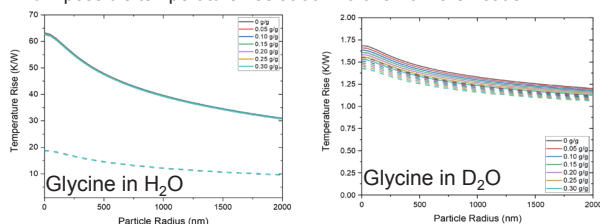


Figure 1: Both; Temperature rise per watt of laser power delivered in solutions aqueous of glycine, using two laser wavelengths 975 nm (solid) and 1064 nm (dashed) at an axial distance of 60 micron from the nearest heat sink while trapping spherical silica particles. Left; Solutions of glycine in H<sub>2</sub>O. Right; Solutions of Glycine in D<sub>2</sub>O.

- Use of D<sub>2</sub>O significantly reduces tweezer induced heating compared to H<sub>2</sub>O.

## Solvent Isotopologue Effects

Induction time distributions were performed to determine the primary nucleation rates response to the deturation of the solvent used [3]. In both solute cases examined, the samples shifted to longer induction time distributions, indicating lower primary nucleation rates.

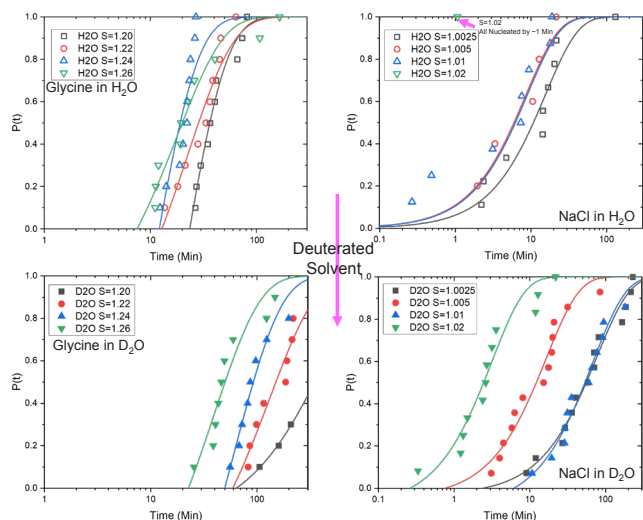


Figure 2: Cumulative frequency distribution of induction times of Glycine and NaCl in H<sub>2</sub>O and D<sub>2</sub>O at the scale of 3 g of solvent performed under agitated isothermal (25 °C) conditions, measured using turbidity. H<sub>2</sub>O shown in open symbols and D<sub>2</sub>O shown in filled symbols. Curves are fitted according to the Jiang and ter Horst model [3]

- At equal supersaturation deuterated solvents give longer induction times under equivalent isothermal agitated conditions.

## References

[1] Yuyama, K. I., Rungsimanon T., Sugiyama, T. & Masuhara, H. Selective Fabrication of  $\alpha$ - and  $\gamma$ -Polymorphs of Glycine by Intense Polarized Continuous Wave Laser Beams. *Cryst. Growth Des.* 12, 2427–2437 (2012)  
 [2] Peterman, E. J. G., Gittes, F. & Schmidt, C. F. Laser-induced heating in optical traps. *Biophys. J.* 84, 1308–1316 (2003)  
 [3] Jiang, S. & ter Horst, J. H. Crystal nucleation rates from probability distributions of induction times. *Cryst. Growth Des.* 11, 256–261 (2011)

## Acknowledgements

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## Impact of Tweezers on Crystal Growth Rates

The nature and magnitude of this impact depend on the solvent isotopologue, presence of a tweezed particle and laser power used. This allows the growth rates to be increased, but this can also prohibit growth in the region closest to the focus of the tweezers, which is evident even after growth was allowed to occur in the region where the laser had prevented it.

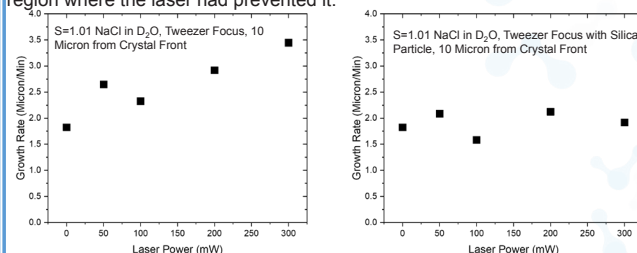


Figure 3: Both; Average of two measurements of Growth rate of a NaCl Crystal in S=1.01 NaCl in D<sub>2</sub>O, when a tweezing focus generated from a 975 nm laser is at an initial distance of 10 micron laterally. Left; Empty Tweezing Focus. Right; Tweezing Focus holding a 1.57 micron diameter silica microsphere.

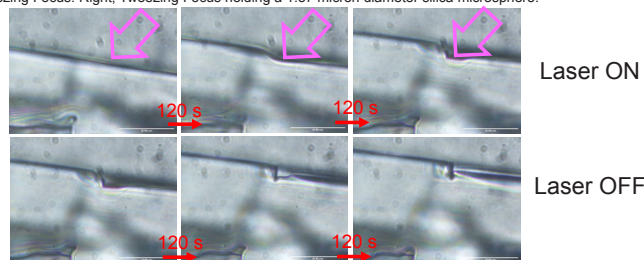


Figure 4: Upper; Growth of a NaCl crystal from S=1.01 solution in H<sub>2</sub>O where the crystal has been allowed to grow through a tweezing focus with a set power of 300 mW. Position of the tweezing focus when active indicated by the arrow. Lower; Further growth following the deactivation of the tweezers.

- Tweezing focuses can increase crystal growth rates in proximity, however this can be negated by the presence of a silica particle, or due to local heating when using non-deuterated solvent and higher laser powers.

## Tweezer Induced Nucleation

Higher laser powers can directly induce nucleation; however, the time distribution over which this occurs depends on several factors, including supersaturation, laser power, polarisation, and solvent isotopologue. However, one factor that has not been previously investigated is the distance between solution-air interfaces and the tweezing focus. It is also possible to induce a crystal to nucleate from what is in bulk undersaturated, which then dissolves back into solution on cessation of the laser.

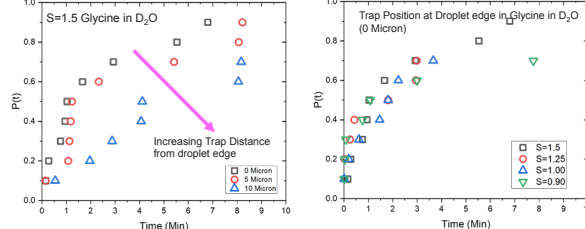


Figure 5: Left; Cumulative frequency distribution of crystallisation times of S=1.5 glycine in D<sub>2</sub>O solution at various lateral distances from the solution droplet edge on the borosilicate glass coverslip using a 1064 nm laser at a power of 770 mW. No nucleation was observed in bulk solution at great distance from the edge. Right; Cumulative frequency distribution of crystallisation times of various supersaturations of glycine solution at the droplet edge using a 1064 nm laser at a power of 700 mW at the focus.

- Tweezing focus distance from the droplet edge alters distribution of induction times, however supersaturations within the levels tested does not have a significant impact on the induction times.

## Next Steps

- Nucleation Time distributions when tweezers are trapping a silica particle.
- Effects of tweezing particles (Silica and Polystyrene) on primary nucleation characteristics.

