

## Supporting Information

# Precise control and measurement of solid-liquid interfacial temperature and viscosity with dual-beam femtosecond optical tweezers in condensed phase

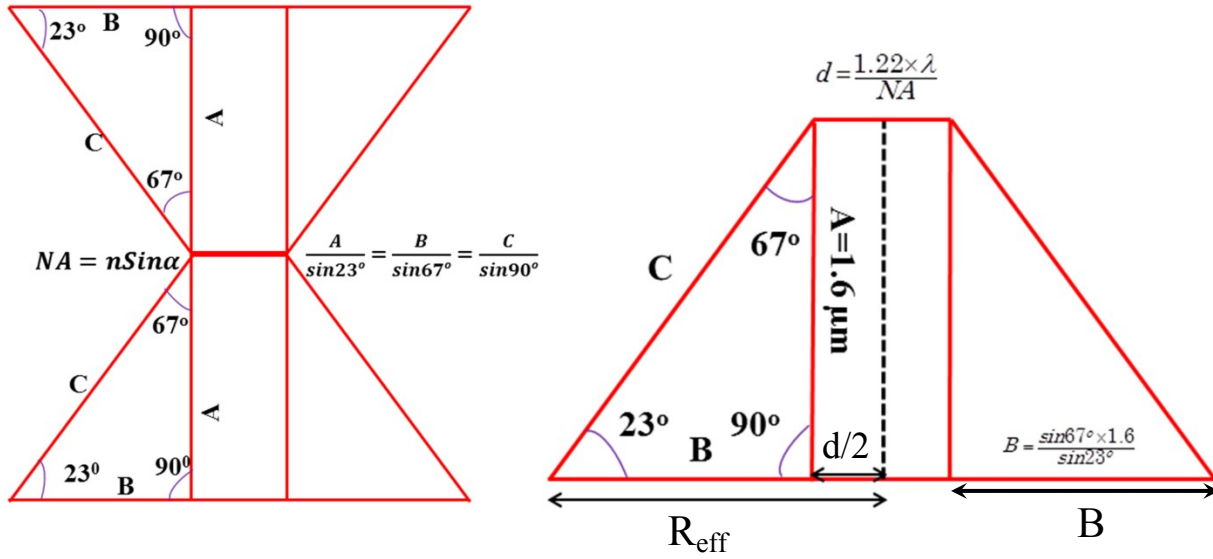
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S1. Model ray optics geometry of optical trapping [Ref. 9 in main article]:



The effective beam waist of 1560 nm at the focus of 780 nm laser where particle is trapped is

$$R_{\text{eff}} = B + \frac{d}{2}$$

$$= \left( \frac{\sin 67^\circ \times 1.6}{\sin 23^\circ} + \frac{1360}{2} \right) \mu\text{m}$$

The effective fluence will be

$$F_0 = \frac{\text{Pulse Energy}}{\pi R_{\text{eff}}^2}$$

$$= \frac{\text{Pulse Energy}}{\pi \left[ \left( \frac{\sin 67^\circ}{\sin 23^\circ} \times 1.6 \right) + .68 \right]^2 \times 10^{-6}}$$

**S2.** The maximum temperature-rise at the interface of the trapped bead and adjacent solvent with respect to room temperature ( $T_0$ ) is:

$$\begin{aligned}
 T_{\max} - T_0 &= 0.783 \frac{F}{\tau_p} \frac{2\sqrt{\tau_p}}{\sqrt{\pi K \rho C}} \\
 \Delta T &= 0.783 \frac{A \times (1 - |r_f|^2) F_0}{\tau_p} \frac{2\sqrt{\tau_p}}{\sqrt{\pi K \rho C}} \\
 &= 0.783 \frac{A \times \left(1 - \left|\frac{n-1}{n+1}\right|^2\right) F_0}{\tau_p} \frac{2\sqrt{\tau_p}}{\sqrt{\pi K \rho C}} \\
 &= 0.783 \frac{A \times \left(1 - \left|\frac{n-1}{n+1}\right|^2\right) \frac{\text{Pulse Energy}}{\pi \left[ \left\{ \left( \frac{\sin 67^\circ}{\sin 23^\circ} \times 1.6 \right) + .68 \right\} \times 10^{-6} \right]^2}}{\tau_p} \frac{2\sqrt{\tau_p}}{\sqrt{\pi K \rho C}}
 \end{aligned}$$

$A$ =Absorbance of solvent used (value given in Table 1 in main article)

$n$ =refractive index of cover glass=1.52

Pulse energy=Average power ÷ repetition rate of the femtosecond pulse laser

Repetition rate of laser=50 MHz

$\tau_p$ =pulse width of 1560 nm heating laser =300 fs

$\rho$ =Density

$K$ = thermal conductivity

$C$  =heat capacity

**S3.** The viscosity of binary mixtures is found as per the following equation (according to Wolf et. al. Ref. 52,53 in main article):

$$\left. \begin{aligned} \ln \eta_{\text{mix}} &= \ln \eta_{\phi}^{\text{id}} + \Delta \ln \eta_{\phi}^{\text{ex}} \\ &= (\phi_1 \ln \eta_1 + \phi_2 \ln \eta_2) + \Delta \ln \eta_{\phi}^{\text{ex}} \end{aligned} \right\}$$

$$\begin{aligned} \Delta \ln \eta_{\phi}^{\text{theo}} &= (\Delta \ln \eta_{\Omega}^{\text{id}} - \Delta \ln \eta_{\phi}^{\text{id}}) + 2g\Omega_1\Omega_2 \\ &= \Delta \ln \eta_{\Omega-\phi}^{\text{id}} + 2g\Omega_1\Omega_2 \\ &= \frac{\gamma\delta\phi_1(1-\phi_2)}{(1+\gamma\phi_2)} + 2\frac{\Delta G^{\text{Rd}}}{RTx_1\phi_2}\Omega_1\Omega_2 \end{aligned}$$

$$\gamma = \frac{F_2/V_2}{F_1/V_1} - 1$$

where,  $F$  and  $V$  represent molecular surface and molar volume respectively, for the component:  $i=1, 2$ . The  $\delta$  can be calculated using relation:  $\delta = \ln \eta_2 - \ln \eta_1$ .  $x_1$ , where  $x_1$  the mole fraction and  $\Delta G^{\text{Rd}}$  is the molar residual Gibbs free energy of mixing at temperature  $T$  (in K). Surface fraction,  $\Omega$ , can be represented by following equation:

$$\Omega = \frac{(1+\gamma)\phi}{(1+\gamma\phi)}$$

All the parameters used in above are calculated from the following Table-I.

Table I: The parameter used at temperature 298 K

Solvent	$\eta$ (Pa.s) $\times 10^{-3}$	F (cm <sup>2</sup> .mol <sup>-1</sup> )	V (cm <sup>3</sup> .mol <sup>-1</sup> )	n	Density (kg.m <sup>-3</sup> )	Mol Wt. (gm.mol <sup>-1</sup> )
Water	0.89	1.8	9.7	1.33	997	18
Methanol	0.53	3.58	22	1.32	786	32
Ethanol	1.06	4.93	32	1.39	785	46

The viscosity of binary mixtures is calculated as a result of temperature rise as follows:

$$\eta_{\text{binary mix}} = \exp \left\{ (0.9 \times \ln(2.414 \times 10^{-2} \times 10^{\frac{247.8}{140-T}})) + (0.1 \times (A_i + \frac{B_i}{C_i + T})) + \Delta \ln \eta \right\}$$

This equation appears in the main article as eq. 5. The Vogel equation parameters: A, B and C in mPa.s are used from Dortmund Data Bank (DDB) as follows:

$$\left. \begin{array}{l} A = -6.7562 \text{ mPa}\cdot\text{s} \\ B = 2337.24 \text{ K} \\ C = 84.0853 \text{ K} \end{array} \right\} \text{Parameter used for MeOH}$$

$$\left. \begin{array}{l} A = -7.37146 \text{ mPa}\cdot\text{s} \\ B = 2770.25 \text{ K} \\ C = 74.6787 \text{ K} \end{array} \right\} \text{Parameter used for EtOH}$$

**I.** In water-Ethanol binary mixture  $\phi_{\text{Water}}=0.9$  and  $\phi_{\text{Ethanol}}=0.1$

**II.** In water-Methanol binary mixture  $\phi_{\text{Water}}=0.9$  and  $\phi_{\text{Methanol}}=0.1$

**S4.**

In the power spectrum, the measured power is only dependent in the interval  $f$  and  $f+df$ . This power measure does not distinguish between  $+f$  and  $-f$  (i.e. it is direction insensitive). In such cases, it is possible to define one-sided power spectral density (PSD) as (Ref. 60 in main article):

$$P_x(f) \equiv \frac{1}{T_{msr}} \left( |\mathcal{X}(f)|^2 + |\mathcal{X}(-f)|^2 \right) \quad 0 \leq f < \frac{T_{msr}}{2}$$

$$= \frac{2}{T_{msr}} |\mathcal{X}(f)|^2$$

$$= \frac{D}{\pi^2 (f_c^2 + f^2)}$$

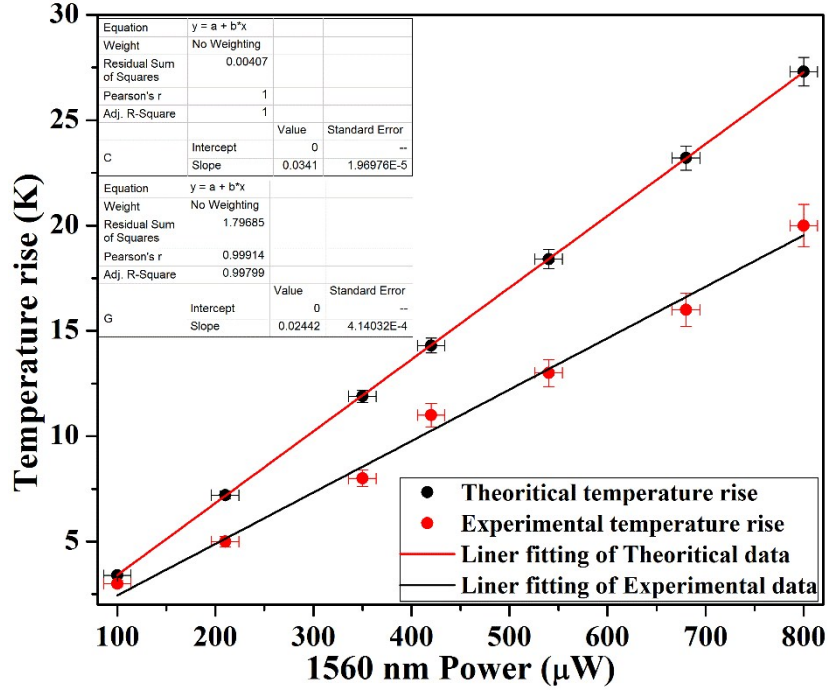
Where  $\mathcal{X}_p(f) = \int_{-T_{msr}/2}^{T_{msr}/2} dt e^{i2\pi f_k t} x(t)$ ,  $f_k \equiv \frac{k}{T_{msr}}$ ,  $k$  is integer and  $x(t)$  is time dependent position taken in QPD with sampling rate 20 kHz.

**S5.** When 1560 nm laser power is absent, the temperature rise is ignored for the optical trap. This is because the temperature-rise is negligible due to small absorbance of solvent at 780 nm. According to room temperature at 298 K, the water-methanol viscosity is 1.30 mPa.s, as calculated from equation 4 in the main manuscript. Experimental data is processed and fitted with equation 7 of the main article to obtain the respective corner frequency within water-methanol at room temperature at 298 K. This leads us to calculate the trap stiffness as:

$$\left. \begin{aligned} \kappa_{TS} &= 2\pi\gamma f_c \\ &= 2\pi(6\pi\eta r)f_c \\ &= 12\pi^2(1.3 \times 10^{-3} \times 0.5 \times 10^{-6} \times 106) \\ &= 0.0081 \text{ pN / nm} \end{aligned} \right\} \quad (\text{Eq - S5})$$

We use the established statement:  $f_c \cdot \eta$  is a constant parameter (Ref 19, 65 in main article). We calculate the local viscosity for individual power of 1560 nm in water-methanol solvent with respect to the room temperature value of:  $106 \times 1.3$  mPa, which gives different corner frequency at different powers of 1560 nm. Now we can get the temperature responsible for individual viscosity by using the equation 4 and equation 5.

S6.



**Figure S6-1:** Temperature-rise as a function of 1560 nm power for both theoretical (black circle) and experimental (red circle) data in pure water and their respective linear fitting in complementary color (Ref. 9 from main article)

We calculate the difference between the gradient of temperature rise,  $\left(\frac{dT}{dP}\right)_{Theo} = 0.034 \text{ K} / \mu\text{W}$ , in theoretically calculated case and the gradient of temperature rise in the experimentally measured case,  $\left(\frac{dT}{dP}\right)_{Exp} = 0.024 \text{ K} / \mu\text{W}$ , for pure water from Fig. S6-1. We use this difference as the correction term that we have now introduced for binary solvents in the following equation 8a (in our main article):

$$T_{\max} - T_0 = 0.783 \frac{F}{\tau_p} \frac{2\sqrt{\tau_p}}{\sqrt{\pi K \rho C}} - \left[ \left(\frac{dT}{dP}\right)_{Theo} - \left(\frac{dT}{dP}\right)_{Exp} \right] C_1 \times P_{avg} \quad (8a)$$