Digital Holographic Measurement of Nanometric Optical Excitation on Soft Matter by Optical Pressure and Photothermal Interactions

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Outline

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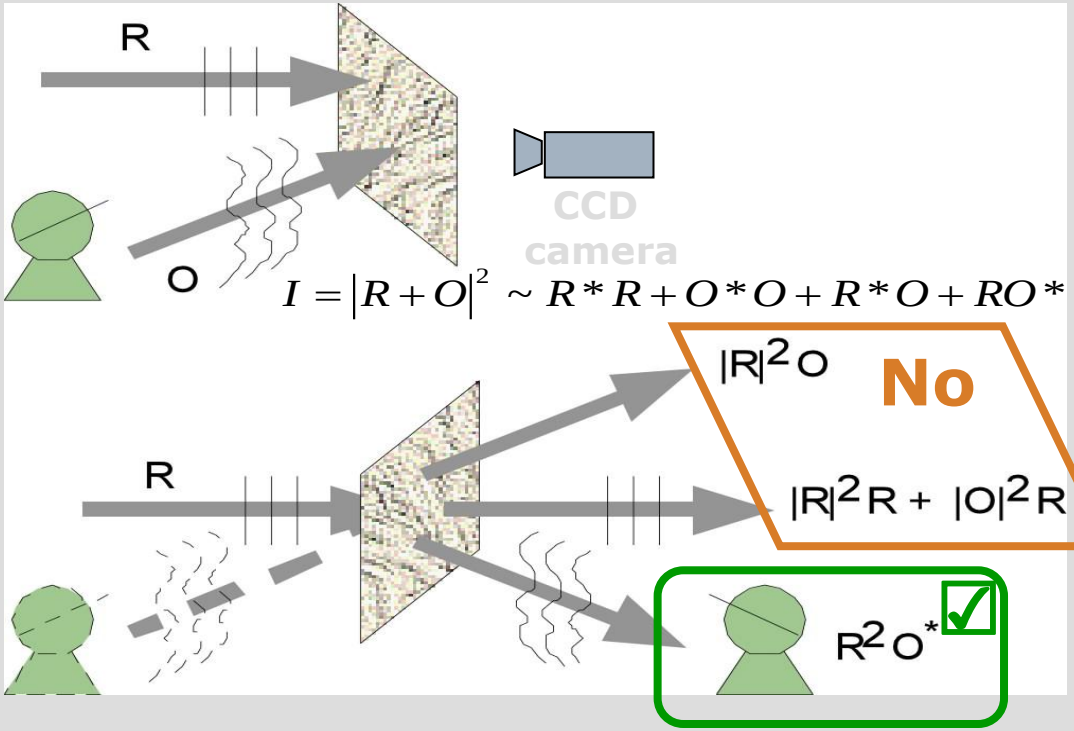
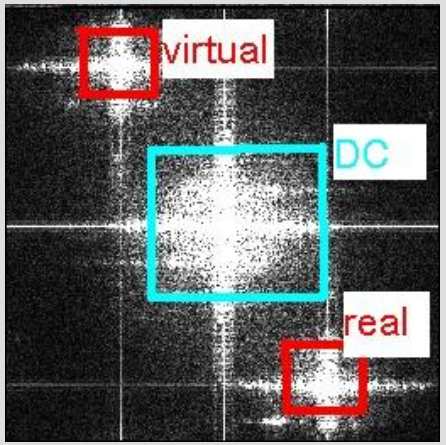
  - DH by the Angular Spectrum Method
  - DH Quantitative Phase Microscopy (DH-QPM) and phase image operations
- Thermal Effects
  - Motivation
  - Thermal Lens Model
  - Overview of Results
- CW Optical Radiation Pressure
  - Motivation
  - Theory
  - Time-dependent Decoupling from Thermal Effects
- Pulsed Optical Radiation Pressure
  - Motivation
  - Results
  - Application to Surface Properties
- Conclusion and Future Direction

Digital holography: the angular spectrum method





Angular spectrum at z = 0

$$A(k_x,k_y;0) = \mathfrak{F}\{E_0(x_0,y_0;0)\} = \frac{1}{2\pi}\iint E_0(x_0,y_0;0)\exp[-i(k_x x_0 + k_y y_0)]dx_0dy_0$$

Optical field at z = 0

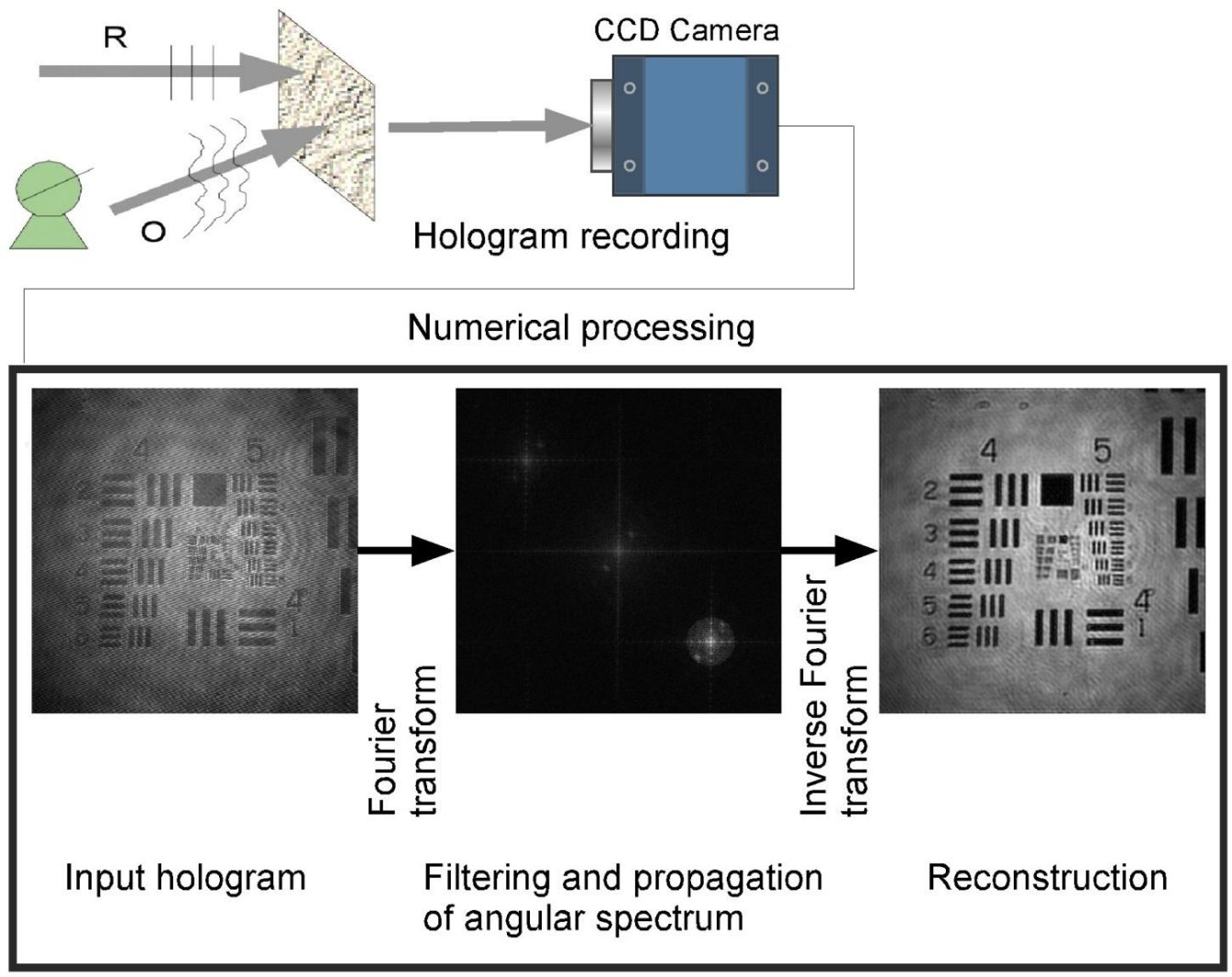
$$E_0(x_0,y_0;0) = \mathfrak{F}^{-1}\{A(k_x,k_y;0)\} = \frac{1}{2\pi}\iint A(k_x,k_y;0)\exp[i(k_x x_0 + k_y y_0)]dk_xdk_y$$

Optical field at z

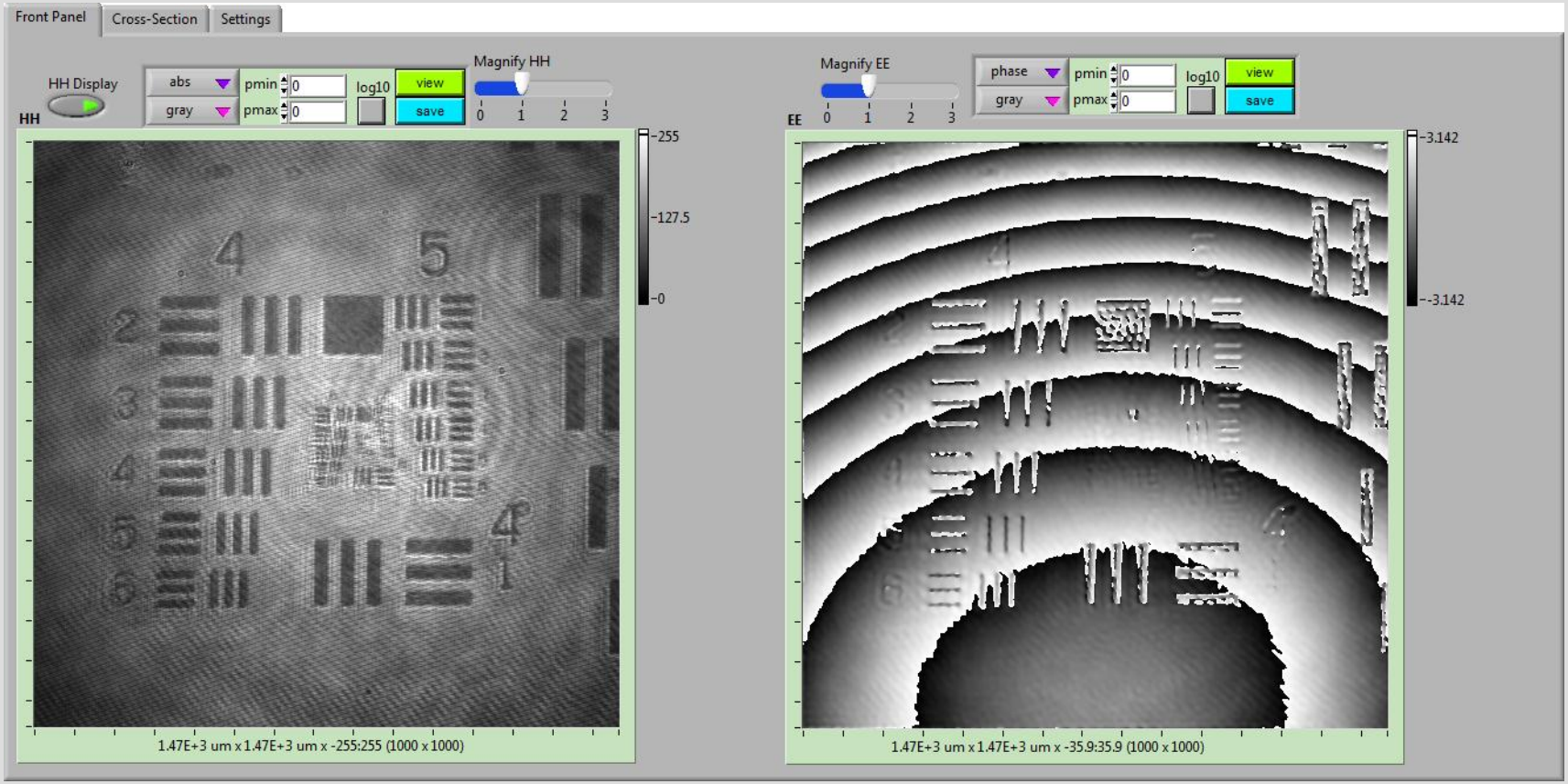
$$E(x,y;z) = \frac{1}{2\pi}\iint A(k_x,k_y;0)\exp[ik_z z]\exp[i(k_x x + k_y y)]dk_xdk_y$$
$$= \mathfrak{F}^{-1}\{\mathfrak{F}\{E_0\}\exp[ik_z z]\}$$

$$k_z = \sqrt{k^2 - k_x^2 - k_y^2}$$

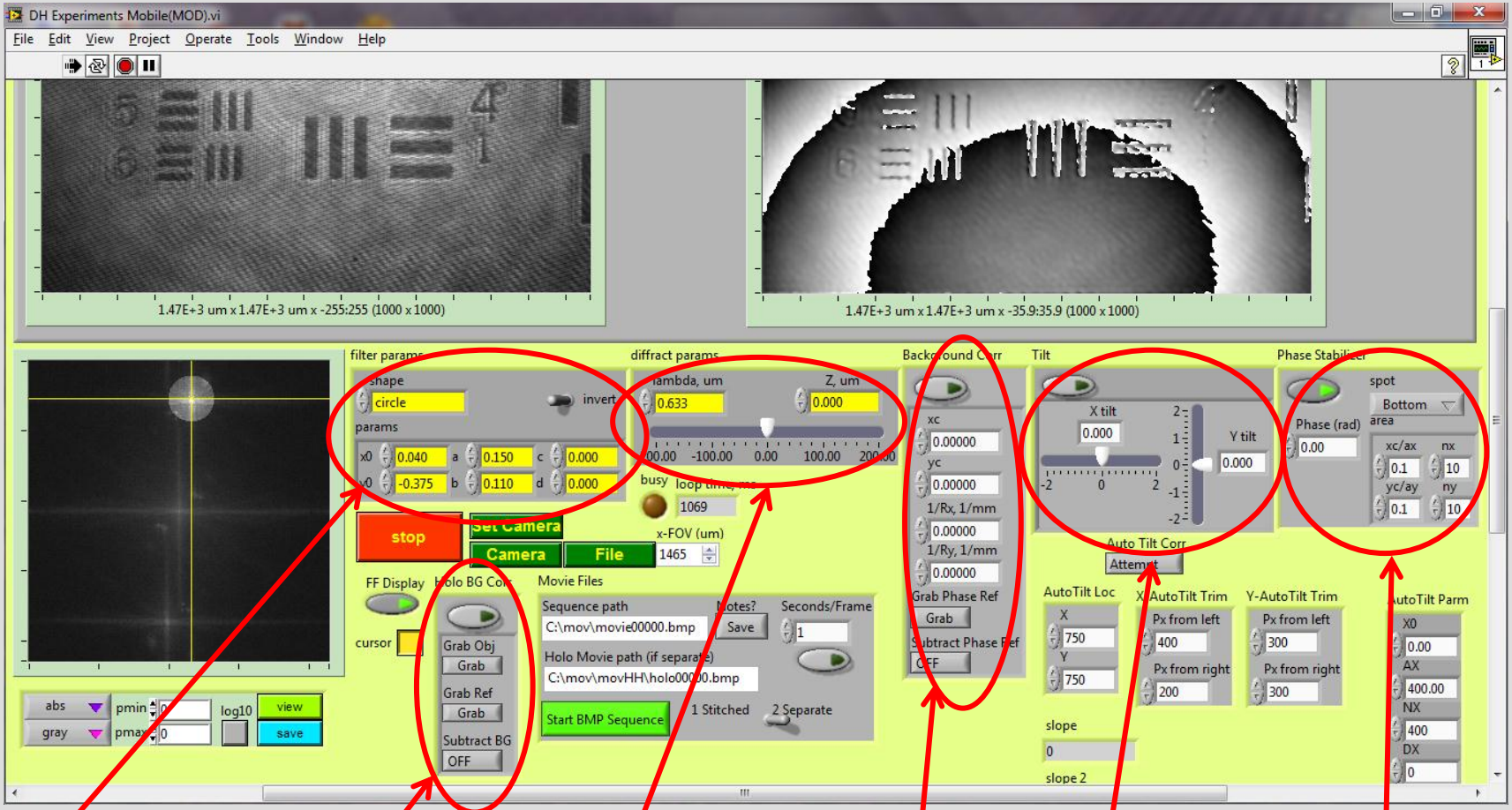
Flow schematic of ASM



Phase imaging: software processing



LabVIEW front panel operations



AS Filter

DC Suppression

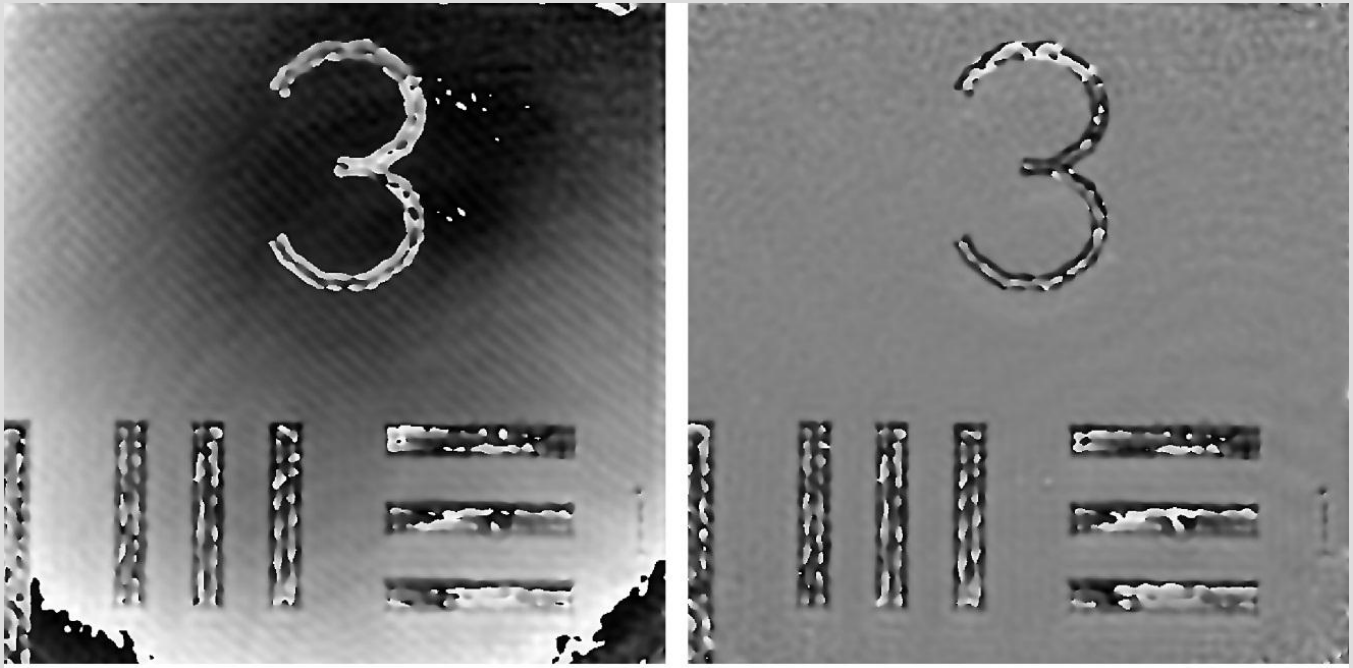
Diffraction propagation

BG aberration compensation

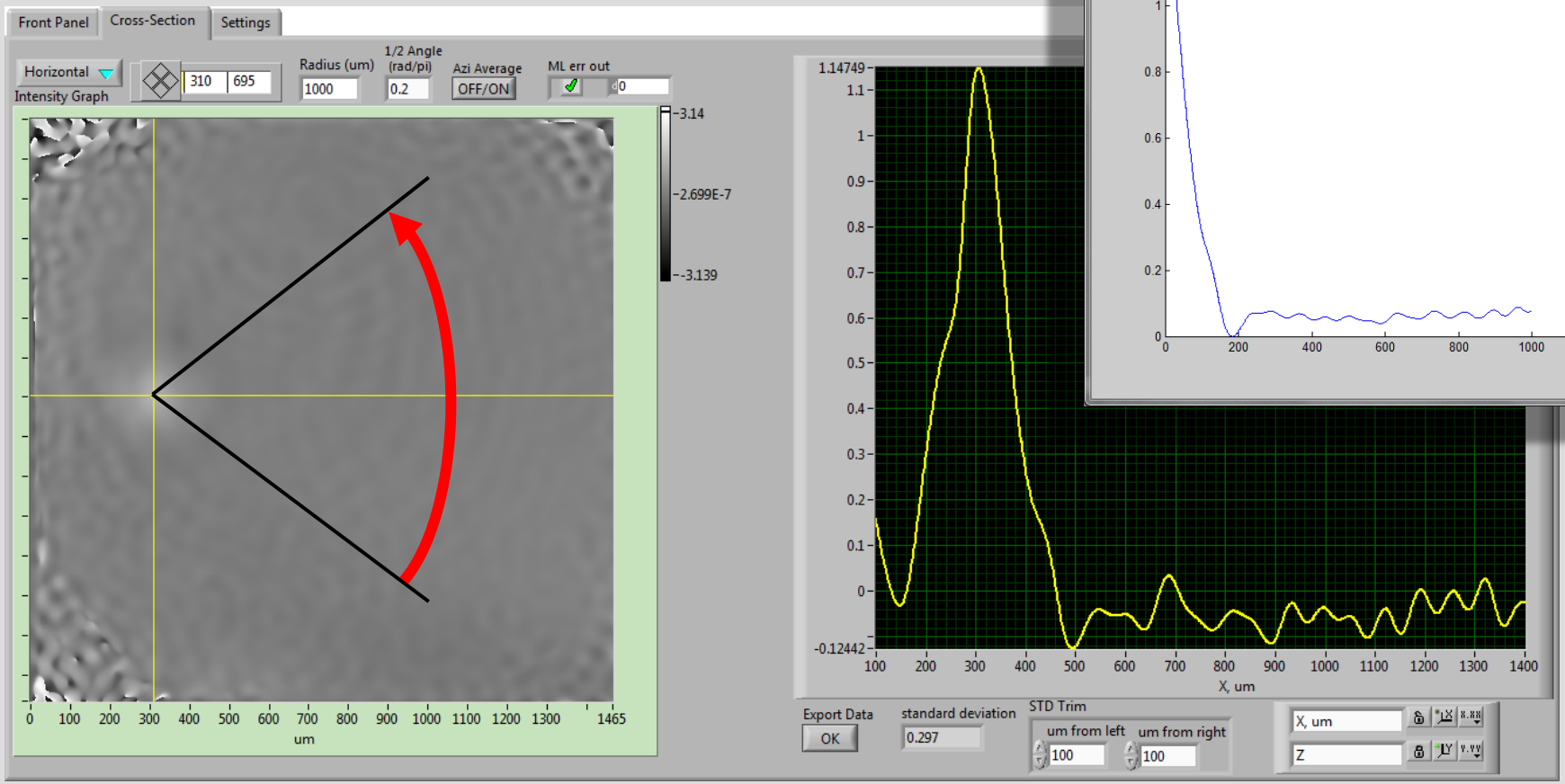
Anti-tilt

Phase stabilization

General aberration compensation



Cross-section operations



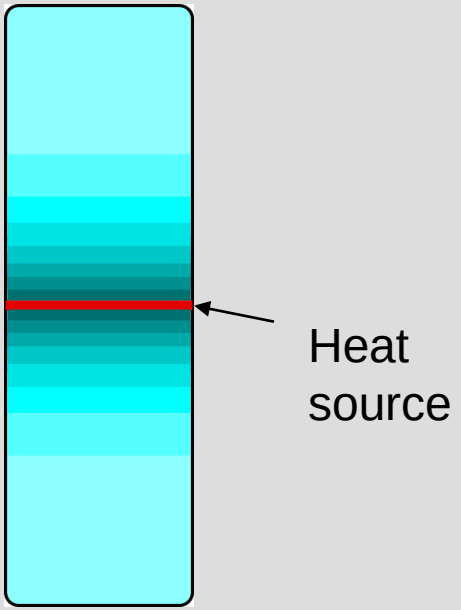
Thermal effects: motivation

- In order to ultimately study the photomechanical effects of optical radiation pressure via DH-QPM, the photothermal effects must be well-understood and characterized.

- Accurate DH-QPM measurement of the thermal lens yields improved measurement of photothermal properties of media.

Thermal effect: thermal lens

VS



Conventional Lens

- n is the same everywhere
- OPL changes because thickness changes

Thermal Lens

- Thickness is the same everywhere
- Cylindrical heat source causes local temperature change
- Thermal diffusion causes temperature gradient
- Temperature dependence of n causes n gradient
- OPL changes because n changes

Thermal lens model: 2D infinite model

Using a Gaussian beam heat source and the solution to the heat transfer equation:

$$\Delta T(r,t) = \frac{2P\alpha}{\pi C\rho w^2}\int_0^t \frac{1}{1+2t'/\tau}\exp\left(-\frac{2r^2/w^2}{1+2t'/\tau}\right)dt'$$

$$n(r,t) = n_0 + \frac{dn}{dT}\Delta T(r,t)$$

$$\phi = \frac{2\pi}{\lambda} l\left[n(r,t) - n(0,t)\right] = \frac{2\pi}{\lambda} l \frac{dn}{dT}\left[\Delta T(r,t) - \Delta T(0,t)\right]$$

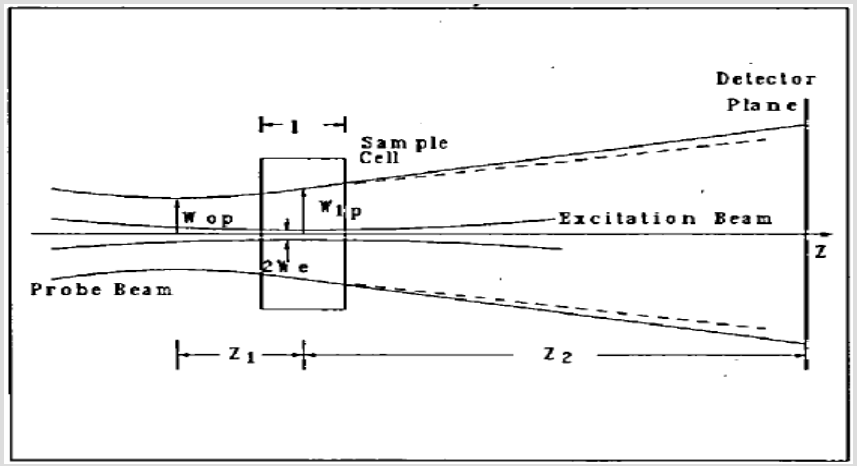
$$\phi = -\frac{P\alpha l(dn/dT)}{\kappa\lambda}\int_0^t \frac{1}{1+2t'/\tau}\left[1-\exp\left(-\frac{2r^2/w^2}{1+2t'/\tau}\right)\right]\frac{dt'}{\tau}$$

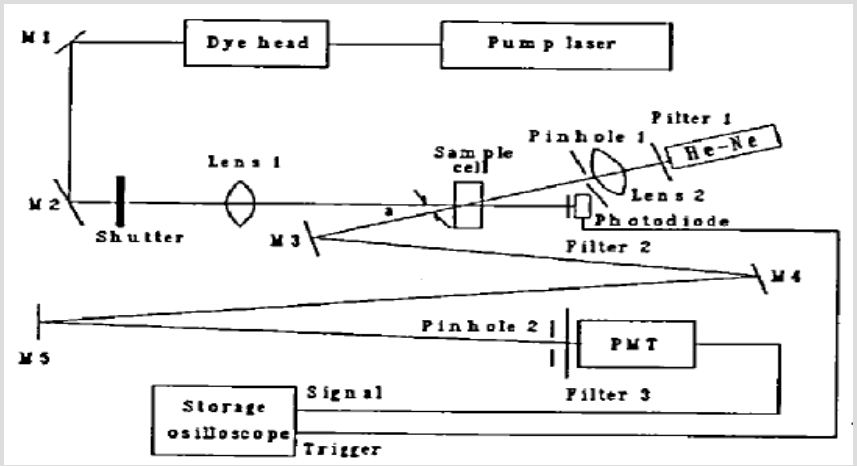
P = pump beam power
α = absorption coefficient
C = specific heat
ρ = density
w = pump beam radius

r = radial distance
t = time
τ = thermal time constant*
κ = thermal conductivity
λ = probe beam wavelength

$$^*\tau = \frac{w^2 C\rho}{4\kappa}$$

The pinhole method of detection





Shen et al (1992)

Disadvantages:
- NOT a direct measure of phase
- Requires assumption of lensing geometry
- Requires long path to detector plane

$$S(z,t)=\Phi_0\,\mathrm{arctg}\{4m(z)v(z)t/t_c(z)/[(1+2m(z)+v(z)^2)2t/t_c(z)+(1+2m(z))^2+v(z)^2]\}, \quad (1)$$

where

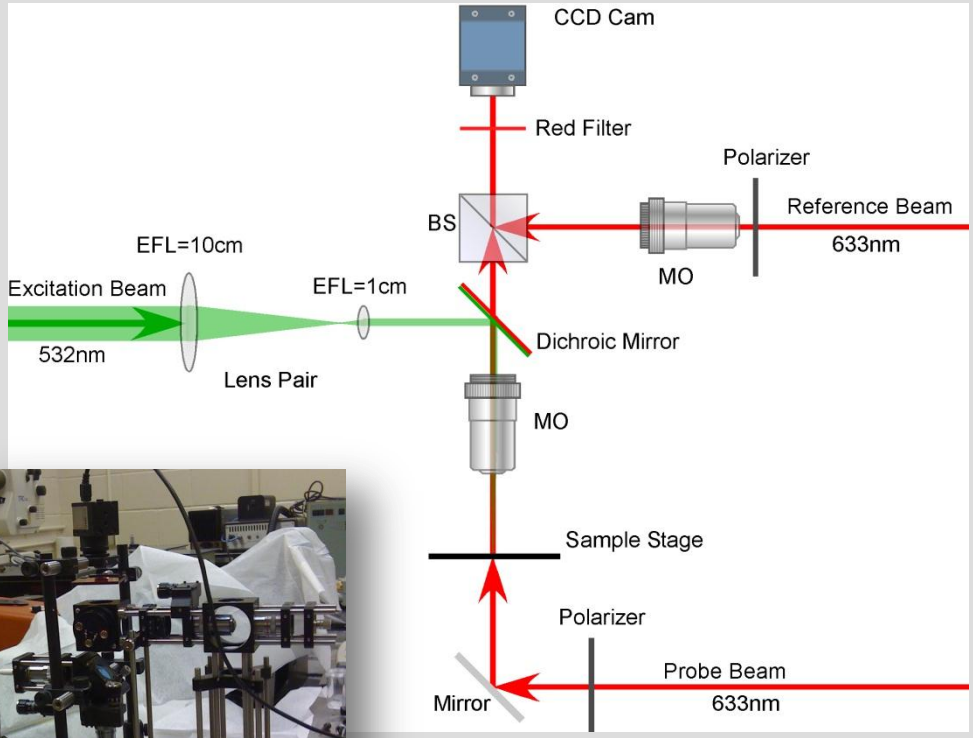
$$m(z)=\omega_p^2(z)/\omega_0^2(z), \quad (2)$$

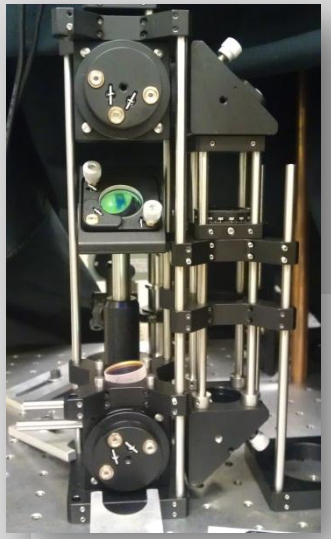
$$v(z)=(z-a_p)/z_p+[(z-a_p)^2+z_p^2]/[z_p(L-z)], \quad (3)$$

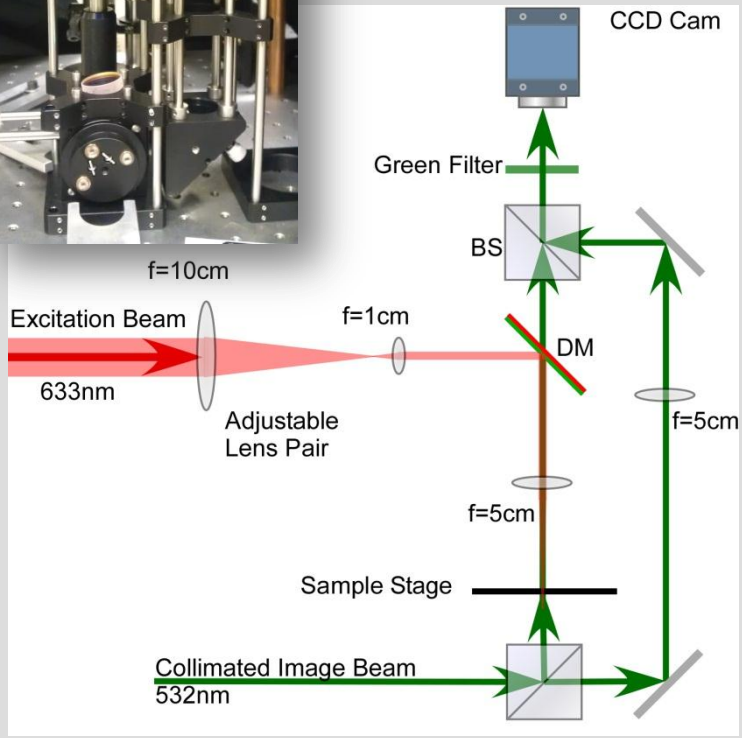
$$\omega_{p,0}(z)=b_{p,0}[1+(z-a_{p,0})^2/(z_{p,0}^2)]^{1/2}, \quad (4)$$

$$\Phi_0=P_0\alpha l(ds/dT)/(\kappa\lambda_p), \quad (5)$$

Experiments: thermal lens apparatus



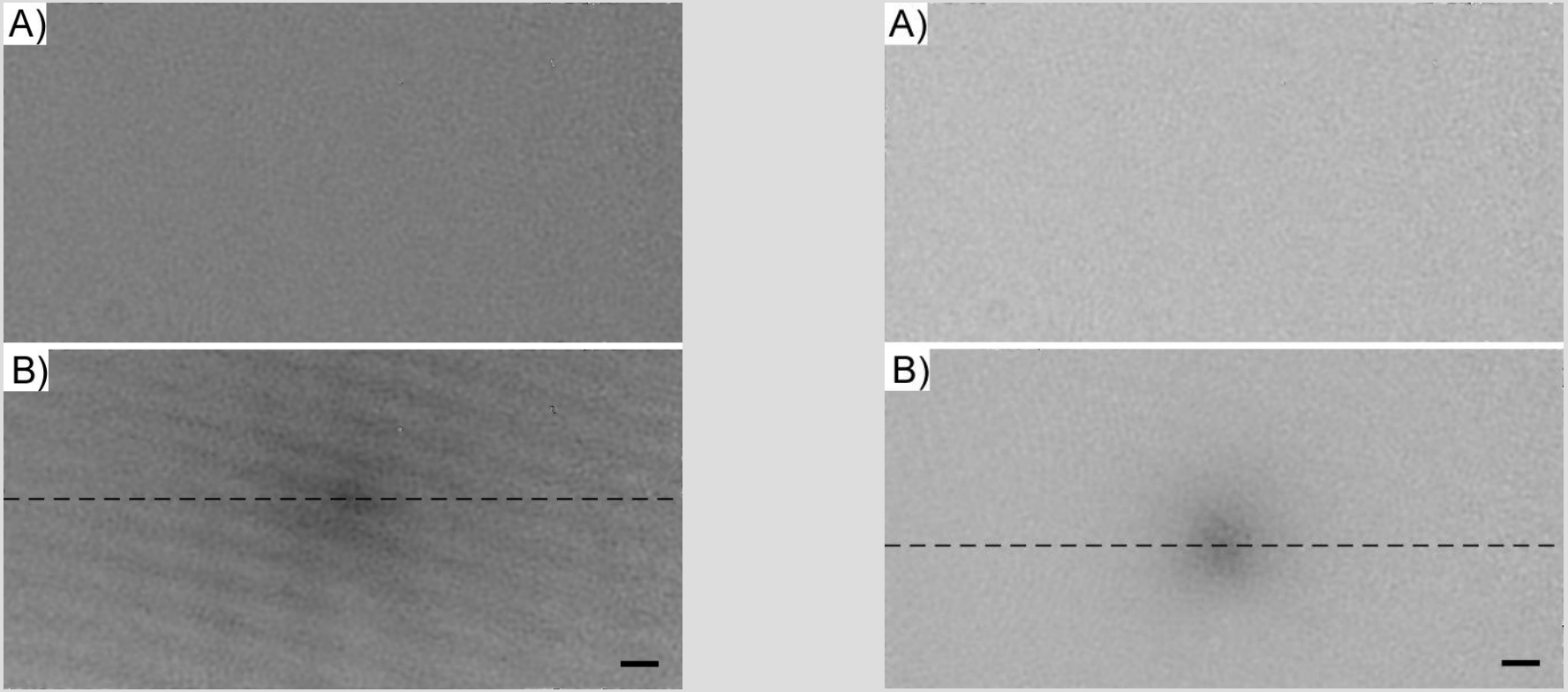




Early Apparatus

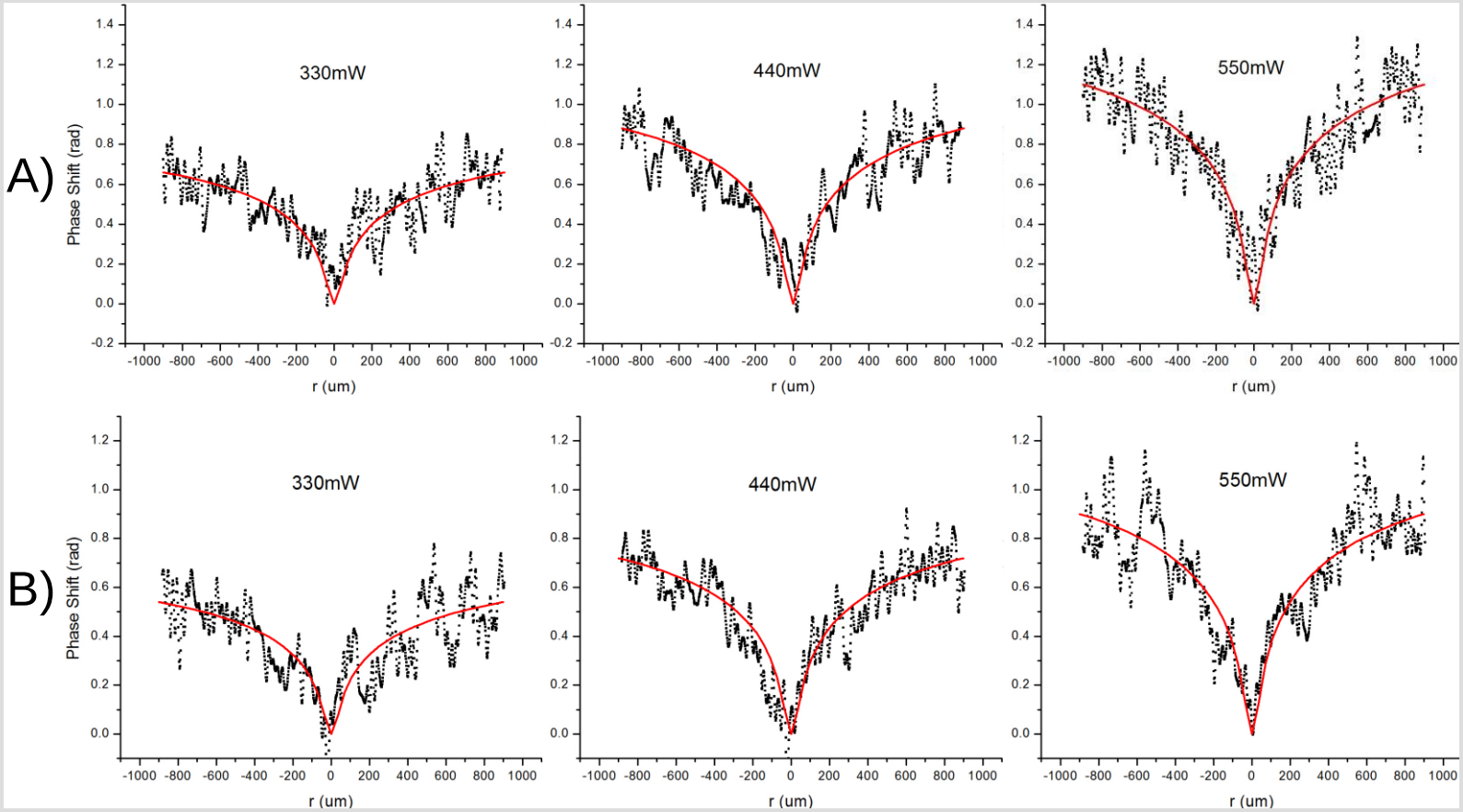
Improved Apparatus

Induced thermal lens phase profile



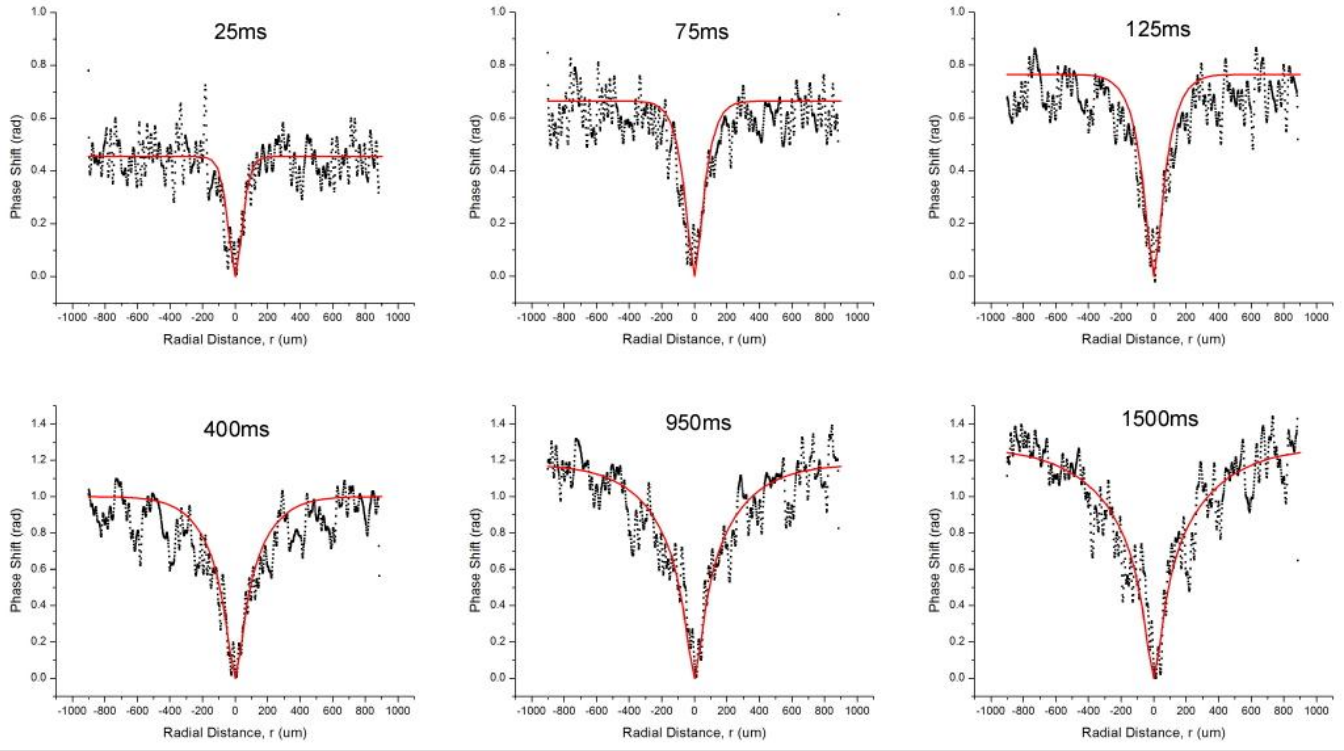
Phase images of a sample, A) with no excitation and B) with optical excitation (thermal lens). Phase scale of images are from 0 (black) to 2π (white) and spatial scale bar is 100μm. Dashed line indicates the selected cross-section

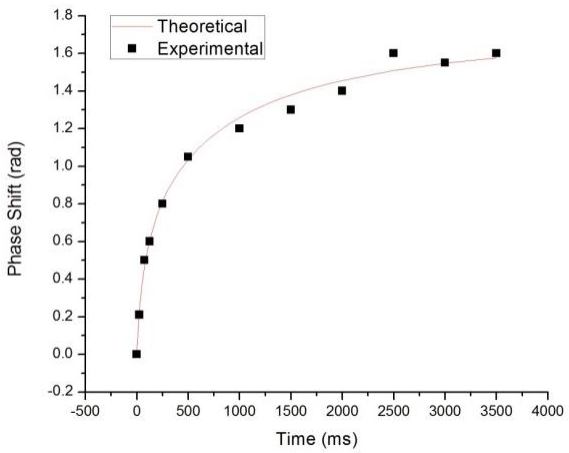
Thermal lens: power dependence



Experimental data (scatter plots) and model predictions (solid lines) at 330mW, 440mW, and 550mW excitation powers for both A) methanol and B) ethanol.

Thermal lens: time dependence





Time-resolved thermal lens phase shift measurements (at *r*=900μm) of benzyl alcohol with model prediction (solid line).

Experimental data (scatter plots) and model predictions (solid lines) at several time-resolved stages of the thermal lens excitation event in methanol. 20fps (50ms/frame), first image time (25ms) is approximated.

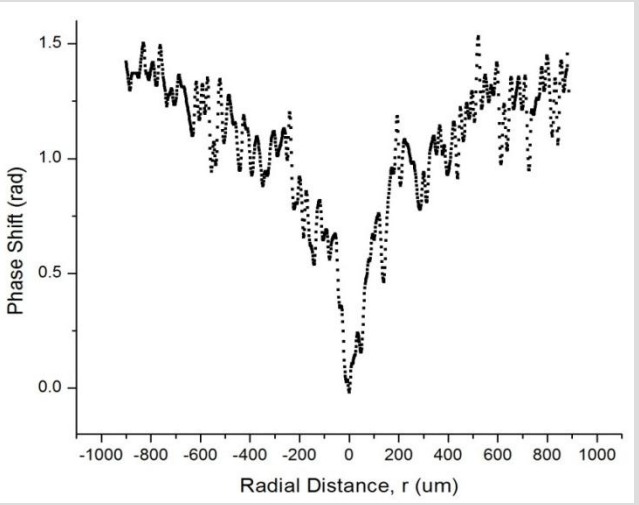
Absorption coefficient

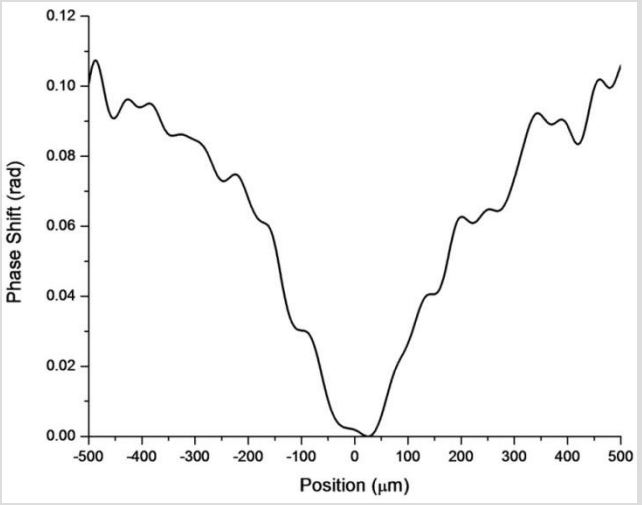
Experimental Parameters and Results for Methanol, Ethanol, and Benzyl Alcohol.

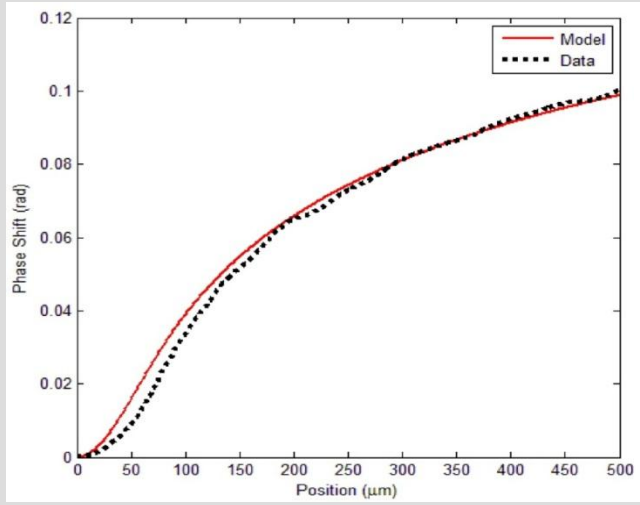
Parameter	Methanol	Ethanol	Benzyl Alcohol**
Power	700 mW	700 mW	30 mW
Excitation Beam Wavelength	532 nm	532 nm	633 nm
Excitation Duration	30 s	30 s	4.000 s
Abs. Coeff. by DH method (cm^{-1})	$(3.6\pm0.3) \times 10^{-4}$	$(2.4\pm0.2) \times 10^{-4}$	$(6.4\pm0.1) \times 10^{-4}$
Abs. Coeff. by PH method (cm^{-1})*	$(5.9\pm0.5) \times 10^{-4}$	$(6.8\pm0.5) \times 10^{-4}$	N/A

*Pinhole method data from Cabrera et al. (2006).
**Benzyl Alcohol study performed with improved apparatus

Summary of improved measurement precision







Parameter	Initial DH-QPM[1]	Improved Apparatus[2]	Azimuthal Averaging[2,3]
Phase (mrad)	120	10	2
Temperature (K)	0.0072	0.0005	0.0001
Refractive Index	2.4×10^{-6}	1.7×10^{-7}	3.4×10^{-8}
Optical Thickness (nm)	12	0.8	0.2
Absorption Coef. (cm^{-1})	3×10^{-5}	1×10^{-5}	2×10^{-6}

[1]Values taken from methanol study.
[2]Values taken from benzyl alcohol study.
[3]Radial symmetry required and actual precision dependent on original system noise level.

Optical radiation pressure: motivation

- Measurement can be useful in soft matter physics for the characterization of fluid interfaces, surfactants, and membranes.
- Noncontact and noninvasive.
- May be adaptable to biological cell membranes.

Optical radiation pressure

Using conservation of momentum across an interface,

$$N\frac{n_1 h\nu}{c}\hat{z} = N\left(T\frac{n_2 h\nu}{c} - R\frac{n_1 h\nu}{c}\right)\hat{z} + \vec{p}$$

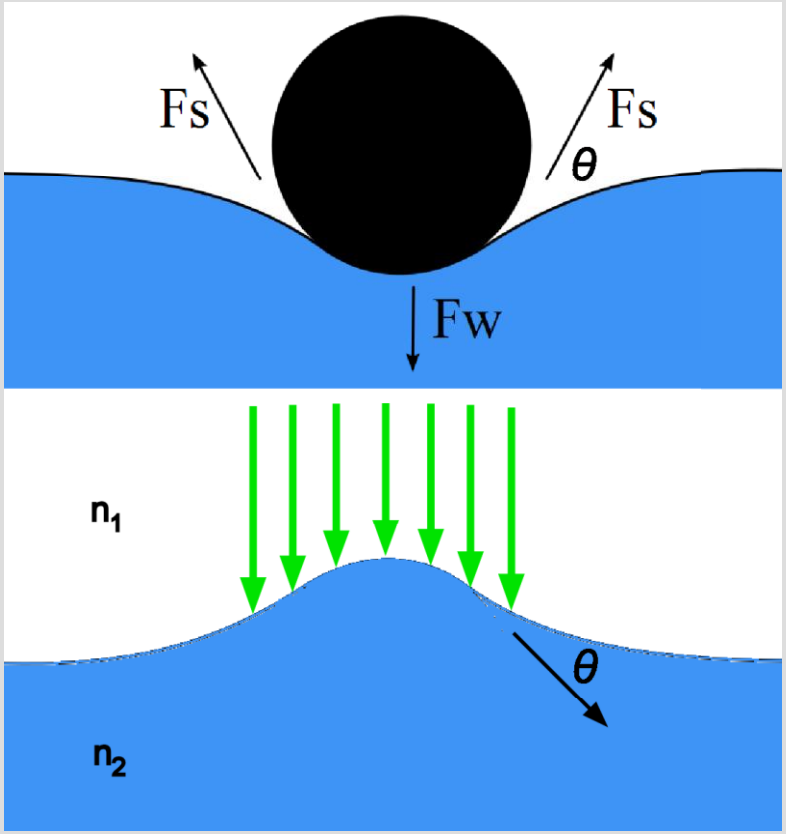
and solving for the simple case of a flat interface with normal incident photons

$$\vec{p} = \frac{2n_1}{c}\left(\frac{n_1 - n_2}{n_1 + n_2}\right)Nh\nu\hat{z}$$

$n_{1,2}$ = refractive index of the first and second media
N = number of photons
T = transmission coefficient
R = reflection coefficient
$\vec{p}$ = momentum transfer to the interface.

** Direction of momentum transfer, and therefore the deformation of the interface, will always point in the direction of the smaller refractive index material regardless of the direction of beam propagation.

CW optical radiation pressure: surface deformation



$$\vec{F}_t = 2\pi w\sigma \sin\theta \hat{z}$$

$$\vec{F}_{opt} = \frac{2n_1}{c}\left(\frac{n_1 - n_2}{n_1 + n_2}\right)P\hat{z}$$

$$\theta = \sin^{-1}\left(\frac{Pn_1(n_1 - n_2)}{c\pi w\sigma(n_1 + n_2)}\right)$$

Optical radiation pressure: deformation height and phase shift

Assuming a spherically growing deformation,

$$h(r) = h_0 + \frac{w}{\sin\theta} - \sqrt{\frac{w^2}{\sin^2\theta} + r^2}$$

with maximum height of deformation (at r=0),

$$h_0 = \frac{w}{\sin\theta}\left(1 - \sqrt{1 - \sin^2\theta}\right)$$

Phase shift associated with deformation,

$$\phi = h\left(n_1 - n_2\right)\frac{2\pi}{\lambda}$$

Time scale of CW optical pressure deformation

The deformation is complete when the net force is equal to zero as described by:

$$F_{net}(\theta) = F_t(\theta) + F_{opt}$$

This value varies as the contact angle, θ, increases from 0 to it's max value. Now, using the integral definition of average to find the average force:

$$\left\langle F_{net} \right\rangle = \frac{1}{\theta_f - \theta_i} \int_{\theta_i}^{\theta_f} F_t(\theta) - F_{opt} d\theta$$

$$= \frac{1}{\theta_f} \left(\theta_f F_{opt} + 2\pi w \sigma \left(1 - \cos\theta_f \right) \right)$$

Now finding the average acceleration from the relation:

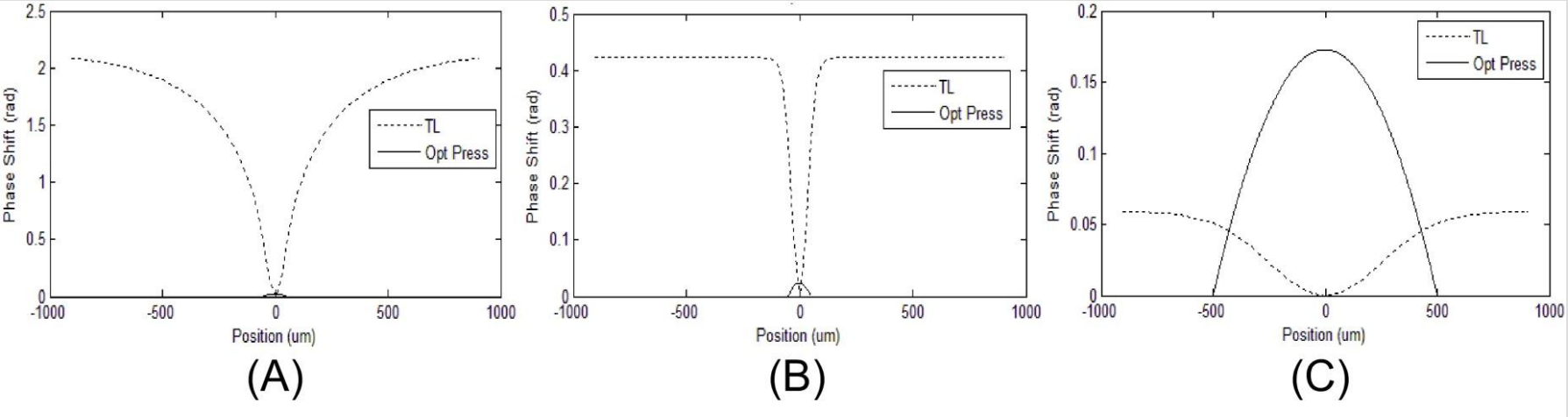
$$\langle a \rangle = \frac{\langle F \rangle}{m}$$

the time scale for the full deformation can be approximated by:

$$t_{opt} = \sqrt{\frac{2h_0}{\langle a \rangle}} = \sqrt{\frac{2h_0\theta_f m}{\left(\theta_f F_{opt} + 2\pi w\sigma\left(1-\cos\theta_f\right)\right)}}$$

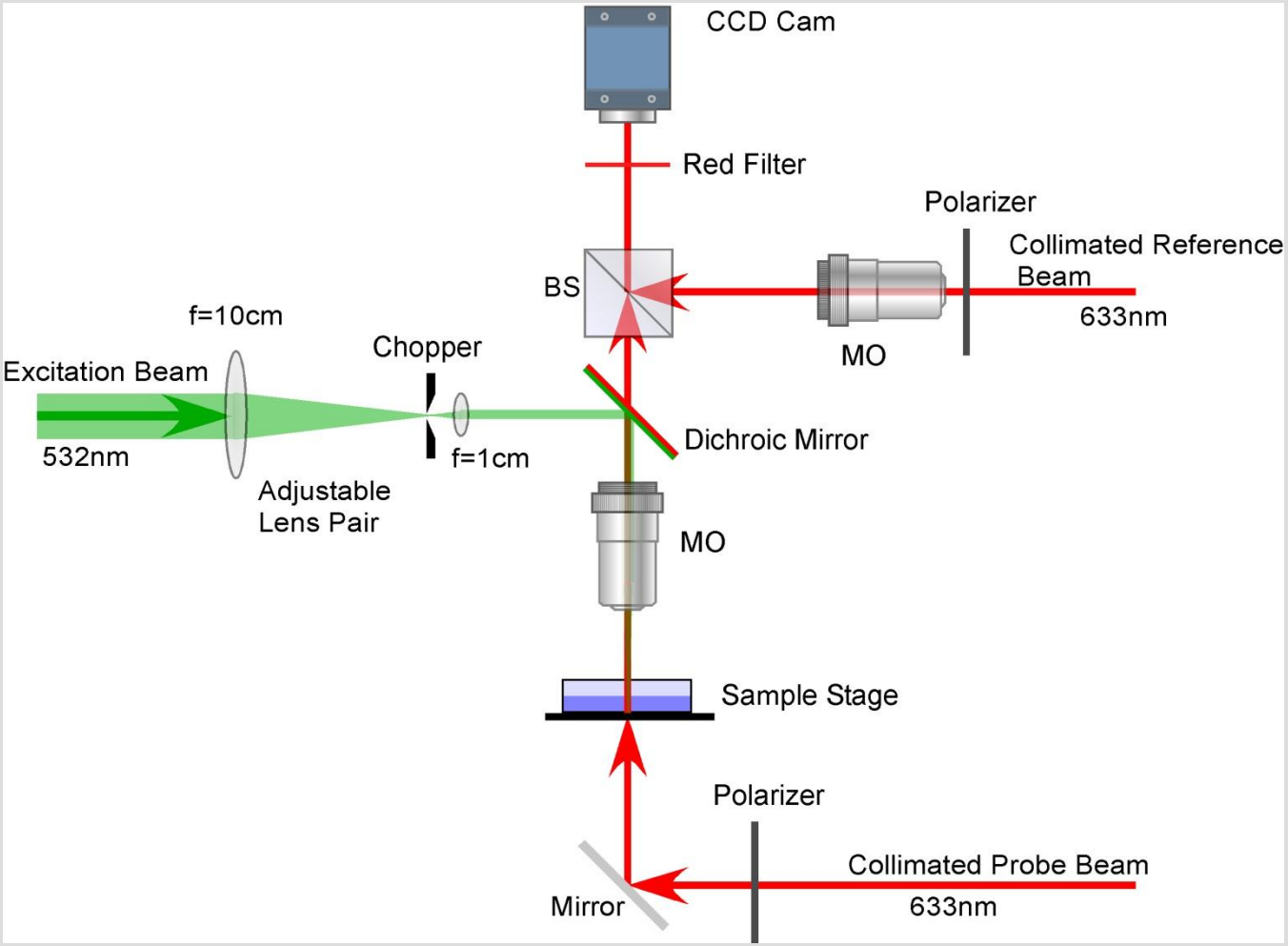
Using the parameters for the upcoming case of water layered on benzyl alcohol and the effective mass of the volume to be shifted, $t_{opt} \approx 8\mu s$.

Simulation of combined model ($BnOH/H_2O$)



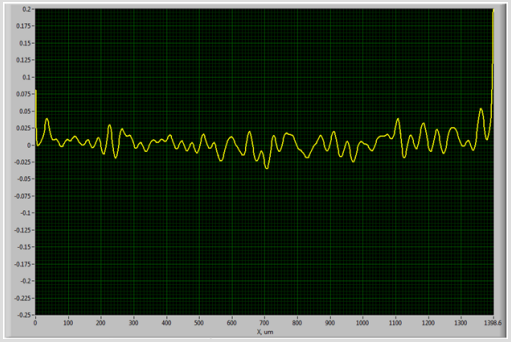
Model prediction for the thermal lens (TL) and optical pressure (Opt Press) effects for a sample of layered benzyl alcohol and water with an interfacial tension of 3.5mN/m. A) 700mW, 50μm beam excitation for a 2 second duration. B) Duration reduced to 10ms. C) 5W, 500μm beam excitation for 10ms.

Experimental apparatus

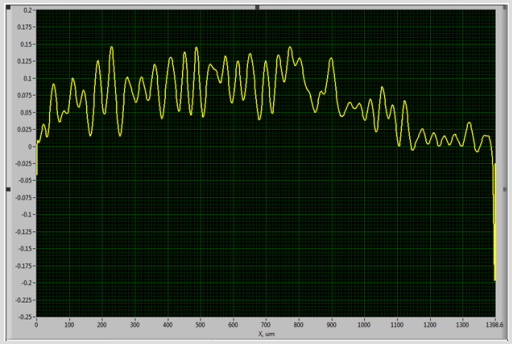


The excitation beam (green) is shuttered by the electromechanical chopper.

Experiment: Optical Radiation Pressure

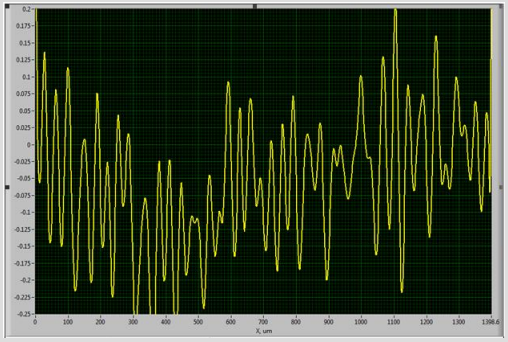


frame0



frame1

frame2



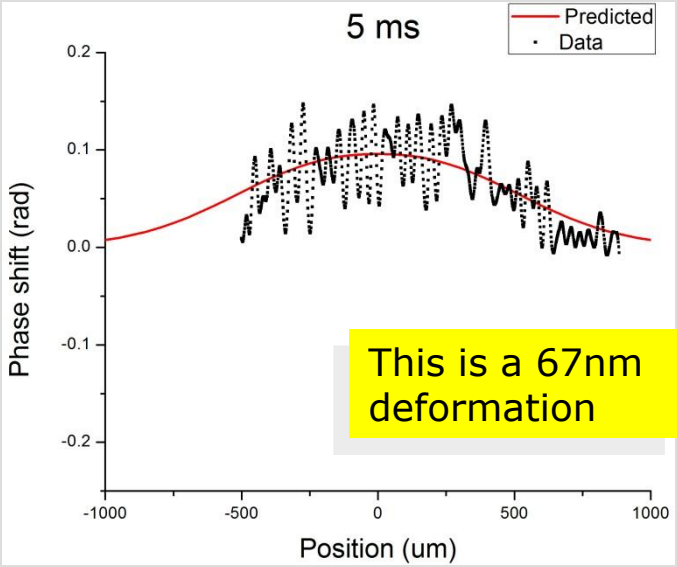
frame3

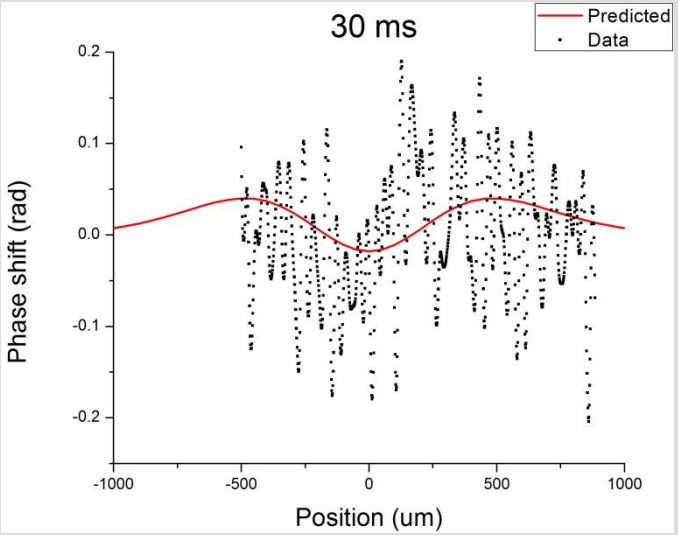
frame4

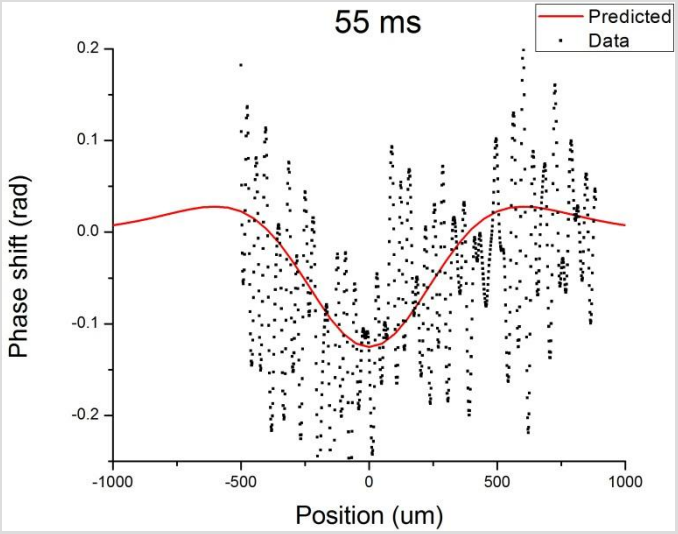
	Water	BnOH
n	1.333	1.540
d(g/ml)	.9971	1.044
κ(W/m/K)	.600	.159
C(J/g/K)	4.181	2.02
τ(ms)	434	829

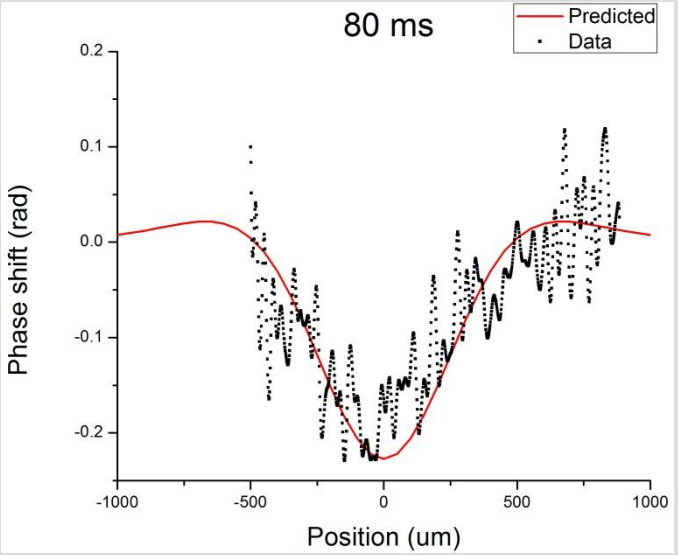
Water layered on top of benzyl alcohol.
Interfacial tension: 3.5mN/m
Excitation beam radius: 500μm
Power: 4W
Camera speed: 40fps (25ms/frame).

Comparing to simulations





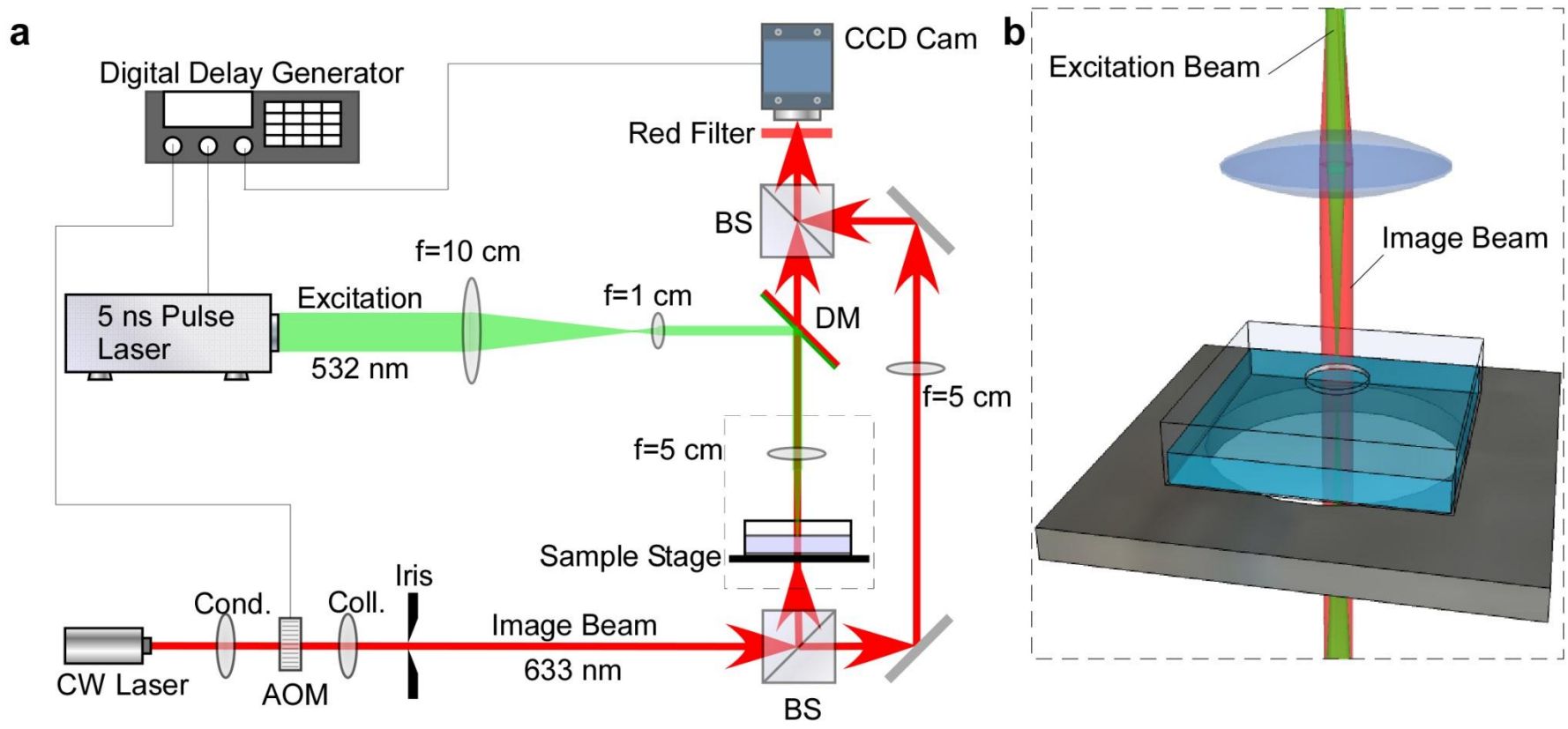




Pulse optical radiation pressure: motivation

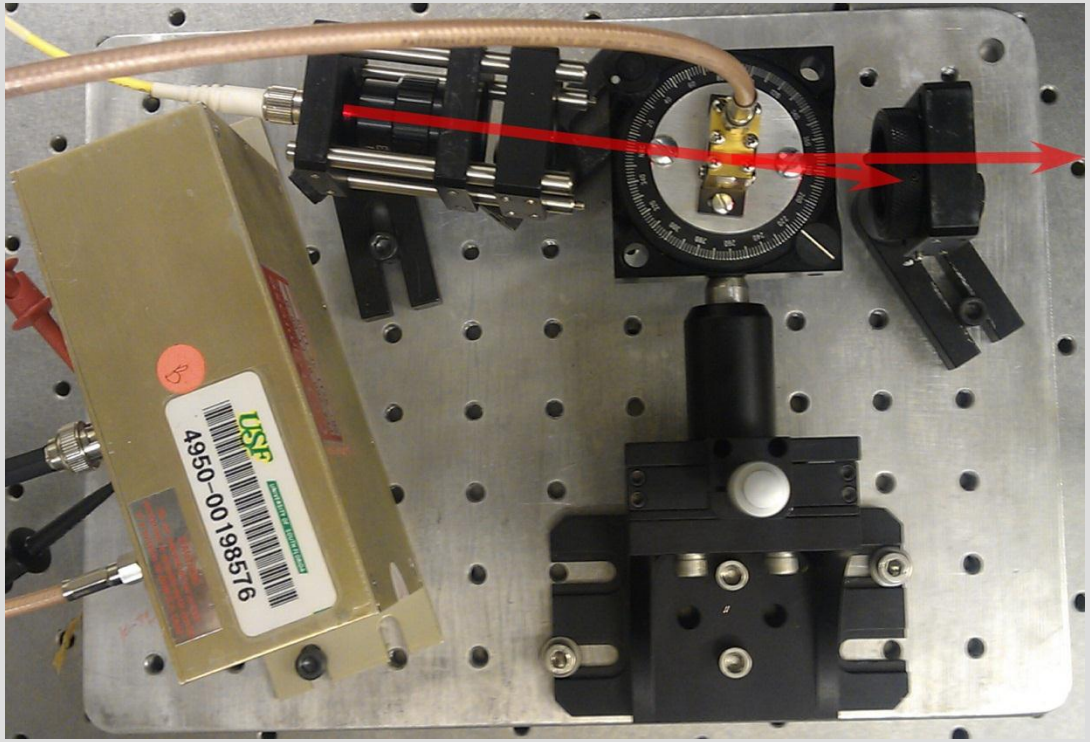
- Reduce thermal effect to negligible.
- CW power available is too weak to deform higher surface tension interfaces such as air/water.
- The time-dependent response of an interface to an optical pressure impulse yields useful mechanical information.

Pulsed optical radiation pressure apparatus

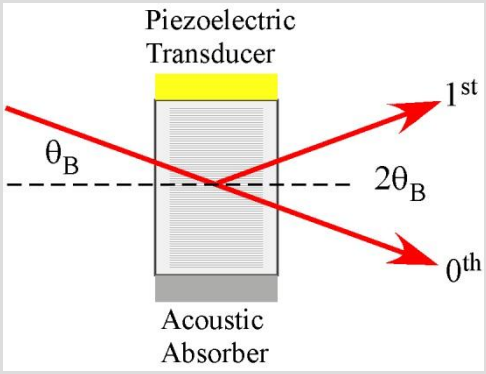


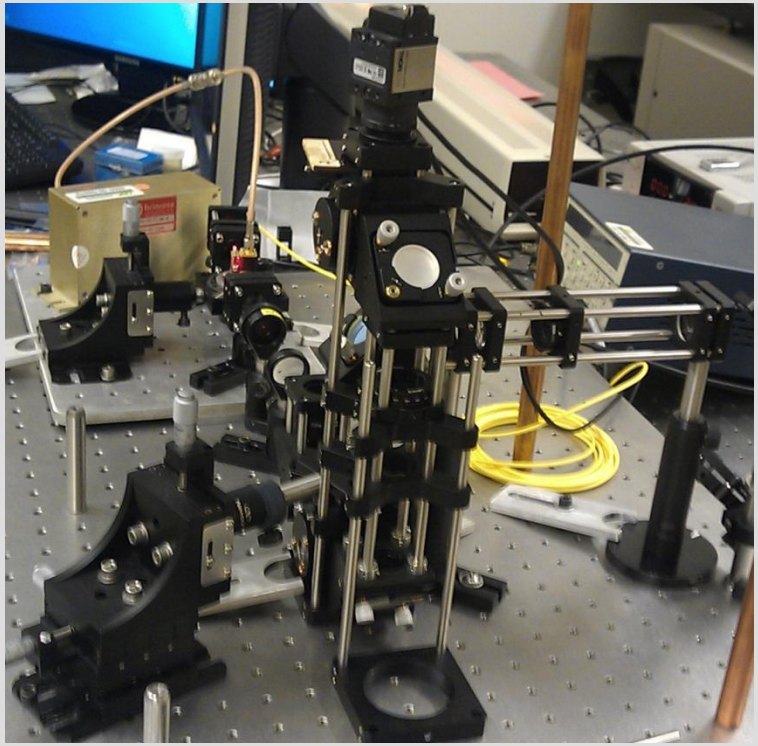
a. CW imaging beam, red, is condensed through acousto-optic modulator, AOM, then collimated through iris (only while AOM is triggered). Collimated beam enters Mach-Zehnder interferometer. Pulsed excitation beam, green, is condensed and collimated by 10:1 focal length lens pair. Dichroic mirror, DM, transmits imaging beam while reflecting the excitation beam down toward sample. Shared 5 cm focal length objective lens loosely focuses the excitation beam onto the interface of interest. **b**. Magnified view of dual-beam sample region (dashed area of a).

Acousto-optic modulator shutter system

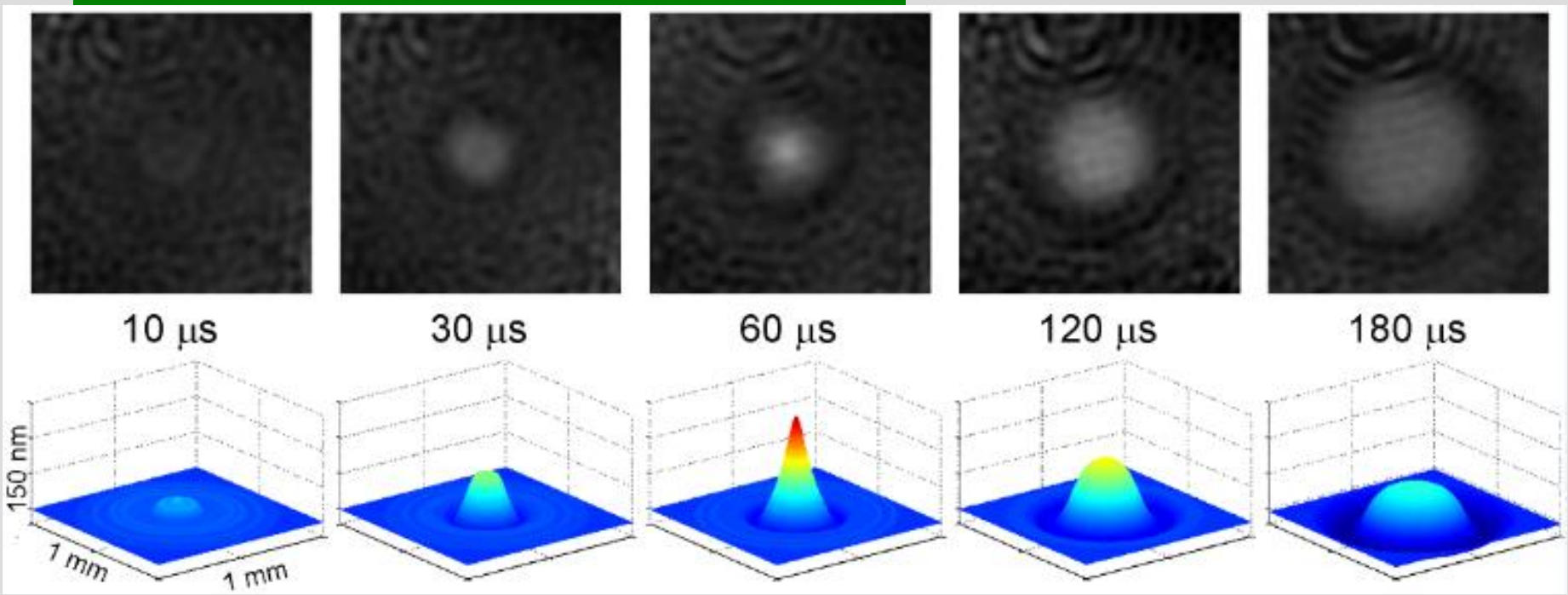


$$\theta_B = \frac{\lambda f}{2V_A}$$

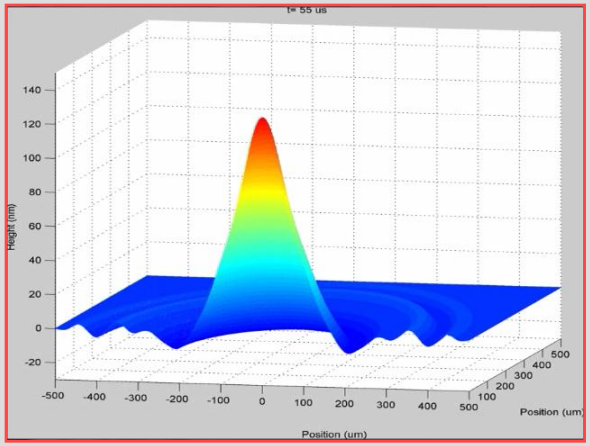




Deformation time-dependence for DI water

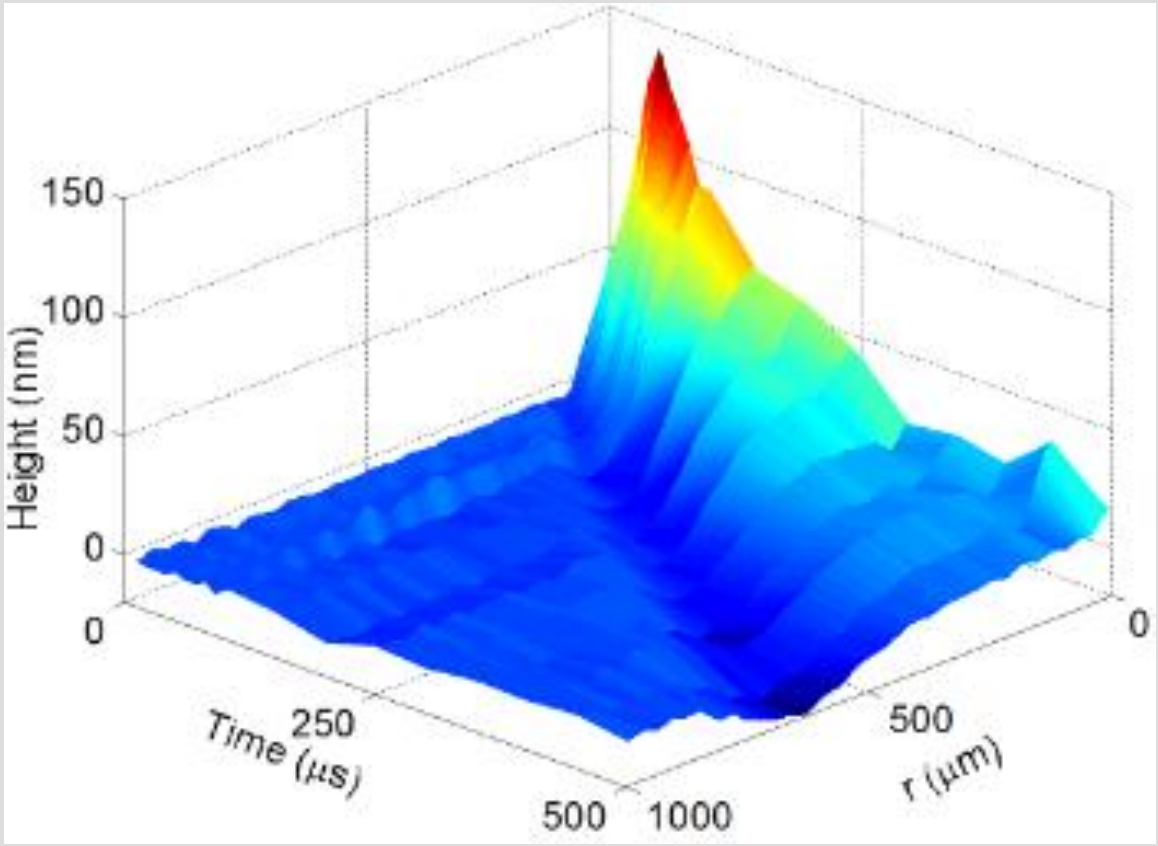


Raw phase images (1.0 x 1.0 mm) of selected time steps and corresponding azimuthal averaged 3D representation (1.0 mm x 1.0 mm x 150 nm).



Total movie time = 300 μs

Time-dependent capillary surface response



Spatiotemporal plot of complete time series.

Capillary wave velocity vs surface tension

The dispersion relation for capillary waves at the interface of two media is described by

$$\omega^2 = \kappa^2 \left(\frac{\sigma \kappa}{\rho + \rho_a} \right)$$

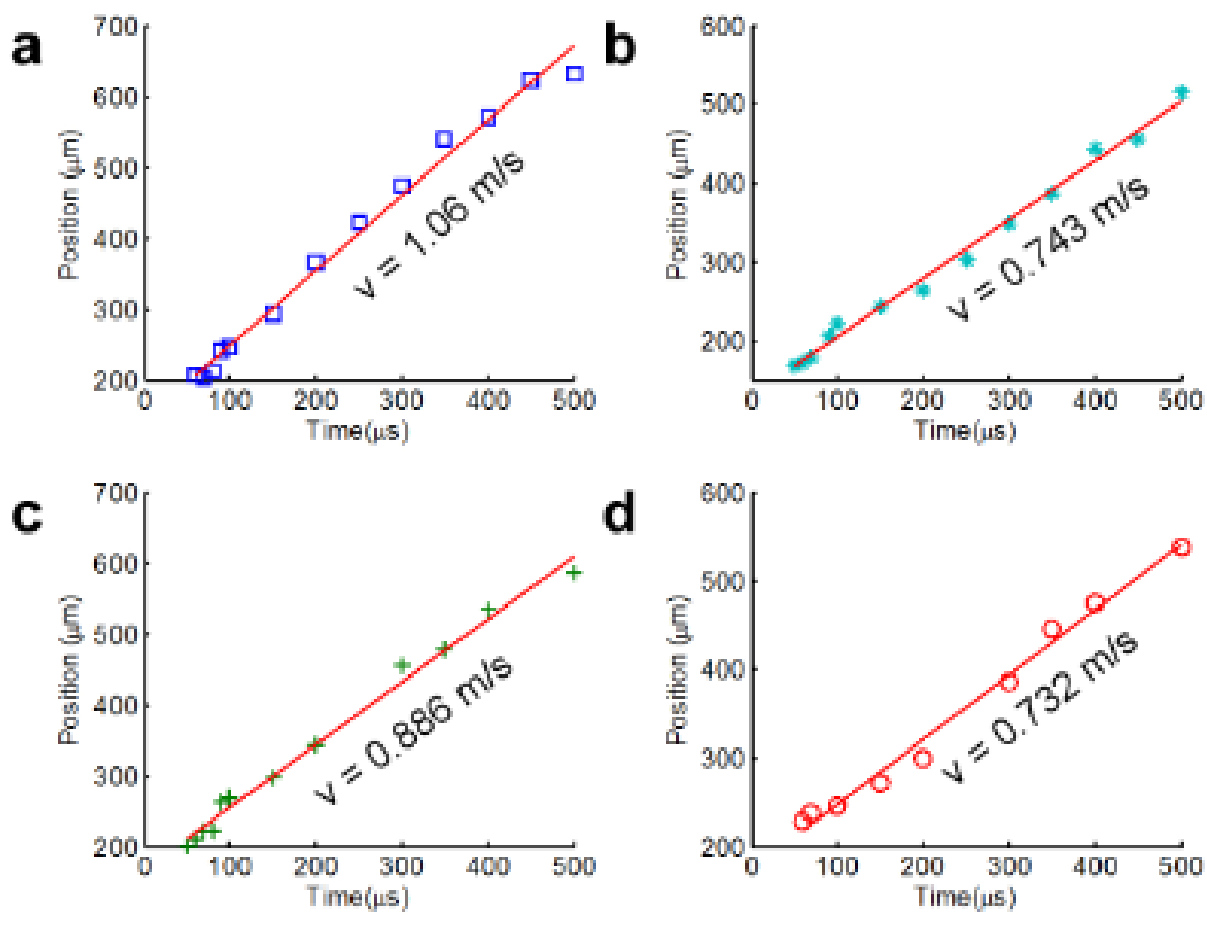
where ρ and ρ_a are the densities of the lower and upper fluids, respectively, κ is the angular wavenumber, and σ represents the surface tension between the fluids. For the present case of the air-liquid interface ρ_a can be dropped since $\rho_a << \rho$ and by substituting the phase velocity, $v = \omega/\kappa$, the relation can be written succinctly,

$$v = \sqrt{\frac{\sigma \kappa}{\rho}}$$

Defining κ at the time step of maximum deformation and integrating with respect to time,

$$r(t) = t \sqrt{\frac{\sigma \kappa}{\rho}} + \frac{\pi}{\kappa}$$

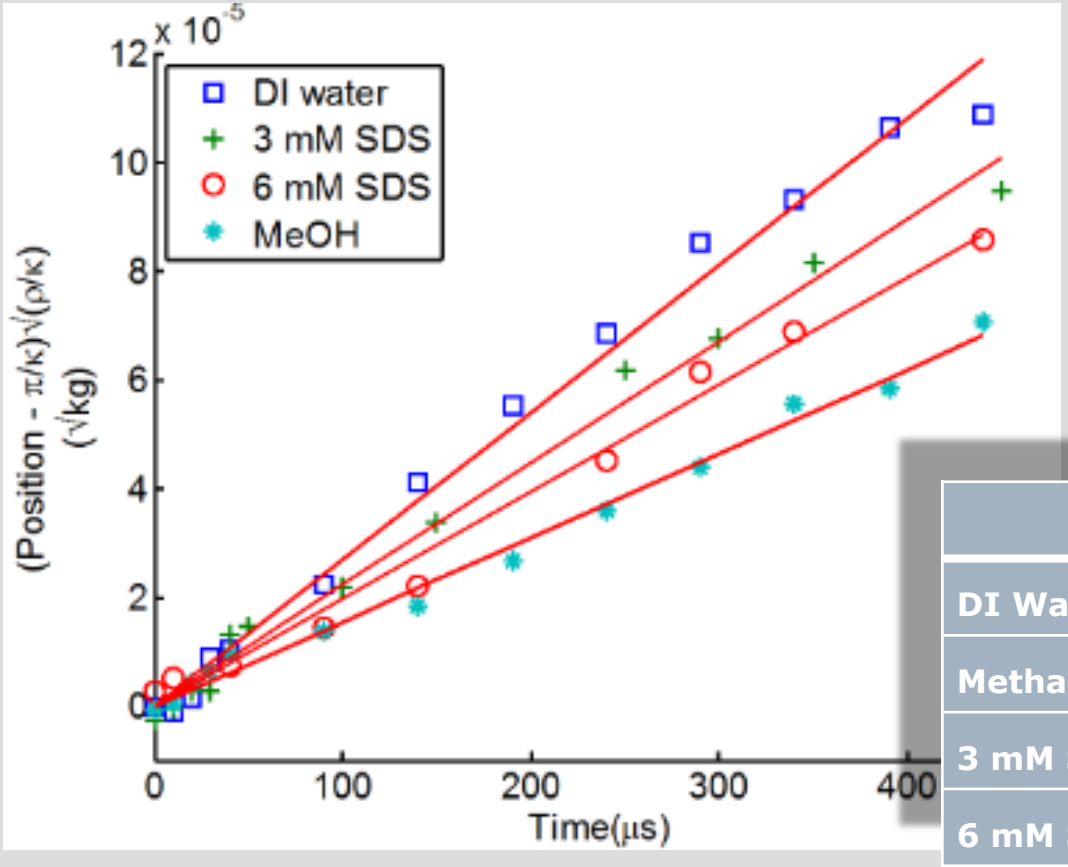
Capillary wave velocity on different surfaces



Position vs. time plots of data and linear fits for: **a**. DI water, **b**. methanol, **c**. 3 mM SDS in DI water, and **d**. 6 mM SDS in DI water.

Comparison plot for air-fluid interfaces

$$\left(r(t)-\pi/\kappa\right)\sqrt{\rho/\kappa} \quad \text{vs.} \quad t$$



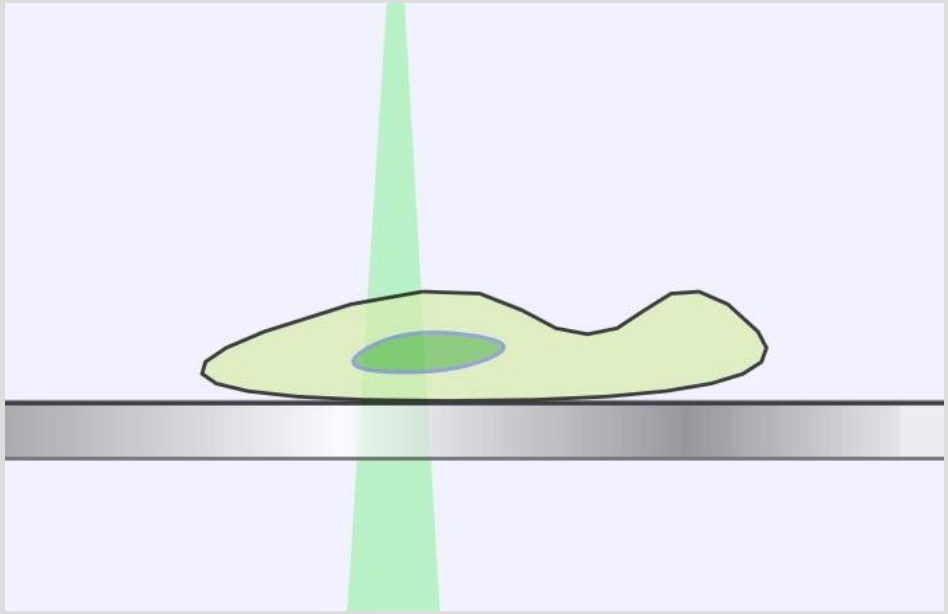
The square of this slope is equal to surface tension (mN/m):

	Published Contact Probe Methods	Noncontact DH-QPM Method
DI Water	72.8	**72.9**
Methanol	22.6-22.9	**23.8**
3 mM SDS	47-57*	**50.1**
6 mM SDS	35-45*	**38.9**

Conclusion

- Demonstrated the capability of DH-QPM to phase image optical interaction phenomena with nanometric and even sub-nanometric precision.
- Realized useful applications for the study of photothermal and photomechanical interactions with fluid media.
- Successfully decoupled thermal effects from optical radiation pressure effects.
- Demonstrated a time-resolved DH-QPM method that can capture and track important surface capillary wave responses without loss of precision.
- Demonstrated successfully a purely optical approach to measure surface energy without contact.

Future direction: application to cell membrane



Requirements:

- Large, mostly transparent cell.
- Cell must behave during experiment.
- Excitation pulse must not damage cell.

Thank you!