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TECHNICAL TUTORIAL

Interactions, Structure, and Microscopic Response:
Complex Fluid Rheology Using Laser Tweezers

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ABSTRACT

Optical trapping techniques are emerging as significant research tools in complex fluids, offering the ability to probe nano- and microscopic interactions, structures, and responses that govern the rheology of complex fluids. In combination with real-space imaging, microstructural response of these fluids can be directly and quantitatively correlated to imposed microscopic stresses and strains. Thus, laser tweezers are enabling us to bridge multiple length scales in colloid and polymer rheology and should be highly useful for investigating the mechanisms of linear and nonlinear rheology. In this article, we briefly review the theory and practice of using optical traps in complex fluids. We discuss the characteristics of the gradient force trap, practical concerns in trapping experiments, and applications, including measurements of micromechanics and microrheology in colloid and polymer gels.

Over 30 years ago, Arthur Ashkin demonstrated that radiation pressure could be used to manipulate individual colloidal particles and cells, levitating them against gravity, or trapping them between counterpropagating beams.[1–3] Shortly thereafter, the first single-beam optical gradient force trap, or “laser tweezer,” was developed, enabling precise, three-dimensional control of particles.[4,5] Optical trapping has since
become a highly effective research tool, particularly in biophysics, by providing a means by which to manipulate cells, organelles, and particles at submicron precision, and to quantify forces from tenths to hundreds of piconewtons. Optical tweezers have played a central role in advancing the knowledge of motor protein mechanochemistry,[6–8] the forces of transcription,[9] and cytoskeletal-membrane interactions.[10] For instance, laser tweezers have been used to measure the stall forces for single molecular motor proteins, such as myosin and kinesin, as they walk along cytoskeletal microfilaments and tubules, in addition to measuring their nanometer step sizes, and the processive movements that drive intracellular transport.[7,11]

Mirroring the development in biophysics, optical trapping techniques are emerging as significant research tools in complex fluids, offering the ability to probe nano- and microscale interactions, structures, and responses that govern the rheology of soft materials. The direct in situ manipulation and force measurements accomplished with optical tweezers are especially powerful in combination with concurrent real-space imaging, such as video, fluorescence, and confocal microscopies. For instance, the pioneering work of Chu and coworkers introduced single-polymer visualization and manipulation to understand polymer dynamics in dilute and entangled solutions of DNA.[12–14] Previous to these works, the physics that underlie many aspects of bulk rheology were only accessible to simulation techniques or scattering experiments. By providing a means of nanoscale force sensing, microscale directed assembly, mechanical measurements, and microrheology, laser tweezers will enable us to bridge multiple length scales to directly establish structure–response relationships in colloids and polymers.

In this article, we briefly review the theory and practice of optical trapping to measure microstructure and response in complex fluids. First, we discuss the underlying theory and characteristics of the gradient force trap. Next, we discuss practical experimental concerns, including common methods for creating and controlling traps. Finally, we discuss several applications, including the use of optical tweezers to measure microstructural response and interactions in colloidal and polymeric materials.

**THEORY OF THE GRADIENT OPTICAL FORCE TRAP**

To generate an optical trap, a single laser beam is focused to a diffraction-limited spot. Two regimes are convenient for describing the physical principles of gradient optical trapping. In the Rayleigh regime, dielectric particles of a diameter much less than the optical wavelength, \( d \ll \lambda \), minimize the energy density stored in the electric field when they are at the center of the focus.[15] Thus, the particle experiences a Lorentzian force from time-averaged electric field intensity pulling it into the light gradient:

\[
F_{\text{grad}} = \frac{1}{2} \alpha \nabla \langle E^2 \rangle
\]  

(1)

where \( \alpha \) is the polarizability of the neutral particle.[16] The particle also experiences a scattering force \( F_{\text{scatter}} \) proportional to the rate of scattering momentum and
absorption. To successfully hold a particle in the propagation direction, the gradient force must exceed the scattering force, $F_{\text{grad}} \gg F_{\text{scatter}}$, otherwise, the particle is pushed along the light propagation axis and out of the trap. The scattering force increases with the particle refractive index contrast relative to the surrounding medium as well as absorption.

In the ray-optic regime, $d \gg \lambda$, a gradient trap can be depicted as individual rays refracting through the particle, as shown in Figure 1. The change in momentum of a photon refracted through the particle imparts a reactive force. For example, in Figure 1, the imparted momentum pushes the particle toward the focal point. By summing the momentum change from all refracted rays, a force profile of the trap can be found. The equilibrium particle position is offset from the beam focus in the direction of propagation due to scattering and absorption. When the size of the particle is on the order of the wavelength, resonant modes between scattering volumes complicate the quantitative description.

Optical tweezers are generally limited to trapping particles with a minimum absorption at the laser wavelength and a relatively low refractive index contrast with the suspending medium. Methods that utilize transverse laser modes other than the Gaussian TEM$_{00}$ are useful for trapping absorbing and highly-reflective particles. In particular, the Laguerre–Gaussian LG$_{03}$ mode, known as the ‘‘donut’’ or ‘‘optical vortex’’ mode, has been used to trap such particles. The LG$_{03}$ mode exhibits a phase singularity along the propagation axis which, by destructive interference, causes the beam intensity to vanish. Laguerre–Gaussian modes can also be used to impart controlled torques onto trapped particles. The magnitude of the torque depends on the topological charge $l$ of the beam. Each photon contributes $lh$ angular momentum. Also, photons in circularly polarized beams carry an additional $\pm h$.

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**Figure 1.** In the ray-optic regime, the gradient force is due to momentum transfer from the refracted beam to the particle, creating a resultant force that, in the case shown, pulls the particle down into the focus. Radiation pressure from scattering and absorption offsets the gradient force in the direction of the light propagation.
As mentioned above, the gradient trap can be used to manipulate and position individual particles. More importantly, for applications in complex fluid rheology, the displacement of the particle from the trap center acts as a sensitive in situ measure of force acting on the particle. This enables us to measure nano- and microscopic responses in complex fluids.

**Trap Compliance and Maximum Trapping Force**

A particle displaced from its equilibrium position in an optical trap experiences a restoring force that pulls it back into the center of focus. The restoring force increases with displacement until the maximum trapping force, or escape force $F_{T,\text{max}}$ (typically from $1–100$ pN) and corresponding displacement $\delta_{\text{max}}$ are reached. At small lateral displacements, the force profile of an optical trap is Hookean; however, the restoring force of the trap becomes nonlinear as the displacement reaches lengths comparable to the particle size. A calculated force profile due to Ashkin\cite{18} is shown in Figure 2A for the ray-optic regime. Although it is difficult to predict the force profile and escape force from the first principles,\cite{26} calculations capture the essential features observed experimentally, most notably the range and nonlinearity at large displacements.\cite{27,28}

The escape force can be estimated from the momentum transfer to the particle $F_{T,\text{max}} \sim Q(nP/c)$, where $P$ is the laser power, $cn$ is the speed of light in a material of refractive index $n$, and $Q$ is a dimensionless trapping force that can reach as high as 0.30 ($Q = 2$ for the radiation pressure exerted on a perfectly reflective plane).\cite{18} While the trapping strength is controlled primarily by the incident laser intensity, particle geometry and refractive index contrast of the suspending medium influence it as well.\cite{19,24}

In the Rayleigh regime, the escape force scales with particle radius $a$ as $F_{T,\text{max}} \sim a^3$, while in the ray-optic regime, it is independent of $a$.\cite{18} Escape forces and displacements compiled by Simmons et al.\cite{27} using a near-infrared laser (Nd:YAG, $\lambda = 1064$ nm) and polystyrene particles with $2a \sim \lambda$ are consistent with Ashkin’s calculations and are reproduced in Figure 2B. They show that $F_{T,\text{max}}$ for trapped

![Figure 2](image-url)
polystyrene particles increases with laser intensity for each particle size. More importantly, $F_{T,max}$ for $2a < \lambda$ increases sharply, then begins to plateau for $2a > \lambda$.

**DESIGN OF AN OPTICAL TRAPPING APPARATUS**

**Beam Steering**

We constructed an optical trap apparatus, shown in Figure 3. We generate two independent optical traps using polarizing beam splitters. The intensity to the traps is controlled by the laser power, while the relative intensity of the traps can be controlled using a half-waveplate. Trap positions in the microscope image plane are controlled by changing the angle of the beam entering the back aperture of the objective. In our dual-trap setup, we use a pair of perpendicular acousto-optic deflectors (AODs, AA Opto-electronic) for the first trap and a motorized gimbal mirror for the second. A telescope consisting of the microscope tube lens and a lens at the side port images the AOD or gimbal onto the back aperture of the objective. To maximize the angular range, we use a telescope magnification close to 1×. Trapping efficiency is maximized by slightly overfilling the back aperture.\(^{[15]}\) For this reason, we selected an AOD pair with large TeO\(_2\) crystals (7 mm), choosing to

![Figure 3](image_url)

*Figure 3.* The optical trapping apparatus. A single laser beam ($\lambda = 1064$ nm) is expanded and split by polarization to form two independent traps. P-polarized beam is steered using a pair of AODs. After recombining the beams, the laser enters the back aperture of a high NA objective. Forward-scattered light can be collected and imaged onto a quadrant photodiode to measure residence times and particle displacement in the trap at fast sampling rates ($\sim 10$ kHz).
expand and collimate the beam before the AOD. However, improved angular range reduces the bandwidth of the AOD to about 100 kHz. The 48 mrad angular range of the AOD enables us to generate an optical trap (or multiple time-shared traps) throughout a $100 \times 100 \mu m^2$ region using our 63× objective, and also enables force clamping (constant stress optical tweezers) by rapidly changing the laser position to maintain a constant separation between the trap and particle centers. The gimbal mirror normally controls the position of a stationary trap, which is used for force measurements. Although steering is generally slow with the gimbal, it is stable over long times. Other beam-steering methods can be used, including translating lenses, galvanometers and piezo-controlled mirrors.\cite{26,29}

Laser

We employ a 4 Watt CW Nd:YAG laser ($\lambda = 1064$ nm), chosen to minimize damage to biological samples, including live cells and DNA.\cite{5} Visible lasers are often used for optical trapping (Ar$^+$ at 488 and 514 nm, frequency doubled Nd:YAG at 532 nm, HeNe at 632 nm), and can be significantly less expensive. For instance, using a visible laser lowers the power requirement and minimizes optical aberrations, because microscopes and objectives are optimized for these wavelengths. Reducing the wavelength also shifts the Rayleigh, intermediate, and ray-optic regimes by a factor of up to two, in theory. In our experience, however, the trapping strength is greatly reduced by power limitations imposed by increased absorption at these wavelengths. For instance, we find that polystyrene particles degrade rapidly at laser powers $\geq 15$ mW at $\lambda = 488$ nm,\cite{28} limiting maximum trap strengths to approximately $F_{T,max} \approx 10$ pN. This is sufficient for particle positioning applications but limits force measurements relevant to complex fluid rheology to fairly small magnitudes. In addition, the traps would no longer be useful for capturing cells or other biologicals, because significant damage results in cell death.\cite{5}

Microscope Objective

To maintain a sufficient axial gradient force to overcome scattering and absorption, it is necessary to use a high numerical aperture objective (NA $> 1$). Oil immersion objectives with numerical apertures as high as 1.4 are available from most manufacturers and are reasonably inexpensive. However, in aqueous samples, the mismatch between the refractive index of the immersion medium and the sample induces aberrations, particularly spherical, that limit trapping to 10–20 $\mu m$ from the coverslip for a 100× NA 1.4 oil objective (Zeiss Plan-Apo). Improved trapping strength and distance from the coverslip can be achieved by using a water immersion objective, such as a 63× C-Apo. These can be highly advantageous when trying to minimize the influence of a nearby interface. If full three-dimensional trapping is unnecessary, two-dimensional traps can be created with inexpensive, low NA objectives. These have been used successfully in total internal reflection microscopy (TIRM) measurements of colloidal forces, to prevent lateral drift of particles and levitate them against gravity with radiation pressure.\cite{30}
Position Detection

A final consideration in laser trapping is position detection of the trapped particle. While it is not a significant concern for pure micromanipulation using optical traps, in situ measurement of forces when probing nano- and microscale mechanical response and interactions is accomplished by measuring the displacement of a particle from its equilibrium position in the laser trap. Two techniques are predominantly used: particle tracking and back focal plane interferometry. The first method simply requires the acquisition of bright field or fluorescence images from the microscope. Particle centers in the image can then be identified to within a subpixel resolution through a series of image-processing operations.[31] Typically, the accuracy of image tracking is \( \sim 30 \) nm, however, time scales are limited by video frequencies (30 Hz). To overcome the spatial resolution and bandwidth of video tracking, back focal plane (bfp) interferometry is a convenient alternative.[32] As shown in Figure 3, laser light from the optical trap is collected using a high numerical aperture condenser. A second lens images the back focal plane of the condenser onto a quadrant photodiode, resulting in an interference pattern that is used to determine the particle position relative to the trap.[33] In addition to increasing the bandwidth to tens of kHz and the spatial resolution to \( \sim 1 \) nm, bfp interferometry allows active control of the trap to generate constant-stress or force-clamp optical tweezers.[8,11]

APPLICATIONS TO COMPLEX FLUID RHEOLOGY

Interactions, Microstructure, and Rheology in Colloids

Micromechanics in Magnetorheological Suspensions

Laser tweezers enable us to measure the interactions and microstructural responses that give rise to bulk rheology in colloidal and polymeric materials. The multiple length scales accessible to laser tweezer experiments enable one to directly associate nanoscale particle interactions to microstructure and macroscopic properties. A clear example is the relationship between dipolar interactions, formation of particle chains, and micromechanics that underlie the bulk rheology of magnetorheological suspensions.[34,35] MR suspensions are colloidal-size paramagnetic particles dispersed in a nonmagnetic fluid. When the dipolar interaction between particles induced by an external magnetic field \( H \) exceeds thermal energy, MR particles aggregate into chains of dipoles aligned in the field direction. The energy required to deform and rupture the new microscopic structure results in the onset of a large, "tunable" yield stress. The ability of MR suspensions to slowly store elastic energy in their microstructures and viscously dissipate it on much faster time scales is common to materials that exhibit yield stress behavior, including electrorheological (ER) suspensions, particulate gels, and foams.[36]

Using laser tweezers, we directly measured the micromechanical properties of individual dipolar chains, as shown in Figure 4. The rupture tensions of chains scaled as \( H^2 \) (Figure 5) are in agreement with the shear stress for dilute suspensions at field strengths below magnetic saturation. In addition, we found that the rupture tensions could be calculated from a self-consistent point-dipole model of the interaction
combined with a repulsive electrostatic double layer. In these studies, a significant increase in microstructural strength was identified due to induction effects and multiparticle interactions along dipolar chains. However, this results purely in an enhancement of rupture tension and does not change the scaling of chain tensile

Figure 4. A chain of 1 \( \mu m \) superparamagnetic emulsion droplets is deformed perpendicular to a magnetic field using two 3 \( \mu m \) “tether” particles. The rupturing angle and tension are dominated by the dipolar interaction between particles. Such microstructural mechanics dominate the rheology of MR suspensions in the presence of a magnetic field and lead to a large, tunable yield stress. The scale bar is 10 \( \mu m \).

Figure 5. Rupture strength versus dimensionless interaction potential \( \lambda = -U_{\text{max}}/k_B T \) for dipolar chains, where \( U_{\text{max}} \) is the maximum attractive interaction between dipoles aligned in the field. The measured rupture tensions are in excellent agreement with calculations based on an attraction between point-dipoles and electrostatic double-layer repulsion (there are no adjustable parameters.) In addition, the linear dependence of the rupturing tension on \( \lambda \) gives rise to the same scaling of the suspension yield stress, clearly linking the interactions, microstructural mechanics, and bulk rheology.
strength with $H$. Thus, it should not affect the field-dependence of the yield stress. In the case of MR suspensions, laser tweezer experiments clearly identified the relationship between nanoscale interactions and microstructural mechanics of polarizable particles to the bulk rheology of MR suspensions.

Assembly and Bending of Gel Backbones

We are currently investigating the relationship between interactions, microstructure, and bulk rheology in other colloidal systems, including particulate gels. Like MR fluids, the rheological behavior of particulate gels ultimately depends on the nature and magnitude of nanoscale interparticle interactions. Attractions induced by van der Waals forces,\[^{37}\] depletion interactions due to nonadsorbing polymers,\[^{38–40}\] and adhesion caused by adsorbing or grafted polymers\[^{41–43}\] cause particles to aggregate into highly branched, tortuous structures.\[^{37}\] Particle interactions, in turn, affect the microstructure, the ability to rearrange, and the tensile strength and bending elasticity of the backbone. At the point at which the microstructure forms a space-spanning network, the bulk rheology exhibits a transition to elastic and yield behavior. We are interested in directly measuring properties that are assumed in models of gel rheology, including the mechanical properties of the gel backbone.

To start, we use time-shared optical traps to directly assemble mimics of gel backbones, as shown in Figure 6. The time-shared traps are generated by rapidly changing the beam angle using our AODs. The scan rate $\nu$ for the laser must be

$$\nu = \frac{2k_BT}{3\pi\eta a^3 b^2} \left[ \text{erf}^{-1} (\gamma) \right]^2$$

(2)

where $b$ is the number of radii a particle is allowed to diffuse, and $\gamma$ is the fraction of particles that must remain within $ba$ of the origin during one scan.\[^{29}\] Glycerol is
sometimes used to decrease the diffusivity of particles,[44] but the high bandwidth of the AOD ensures that we can position and control particles far from the interface, even in aqueous solutions. The trap displacement is then reduced until particles come into contact. At sufficient ionic strengths, van der Waals interactions force the particles into primary minima, creating a rigid chain that mimics the backbone. Note that the chain is formed and held at \( \sim 100 \ \mu m \) separation from the interface to prevent the particles from adhering to the coverslip.

The mechanical properties can then be measured by optical tweezers to establish the bending stiffness of the microstructure that gives rise to macroscopic elasticity in gels.[45,46] An example of one such bending experiment is shown in Figure 7, where 3.14 \( \mu m \) poly(methyl methacrylate) (PMMA) particles have been assembled into a chain in 0.1 M MgCl\(_2\). The chain is held with three time-shared optical traps—two stationary traps at either end and a translating trap in the center. The relative strength of the traps is adjusted so that the center trap has a lower maximum trapping force than the stationary traps. The chain bends as the center trap is moved to the right. A low velocity (\( \sim 0.1 \ \mu m/s \)) of the moving trap ensures that mechanical equilibrium is achieved, and the applied tension can be found by displacement from the stationary traps. The observed deformation is in good agreement with that of a bending rod under an applied load \( F \),

\[
y(x) = \frac{4F}{\pi a^4 E} \left( \frac{Lx^2}{2} - \frac{x^3}{6} \right)
\]  

(3)

for a rod of radius \( a \), length \( L \), and Young’s modulus \( E \). Shown in Figure 7 is the position of particle centers at the point of maximum deformation compared to the equation for a bending rod. After the trap releases, the chain relaxes to the unbent configuration.

We are currently investigating how the mechanics of the gel backbone is influenced by the nanoscale particle interactions. For instance, compare the stiff behavior of chains aggregated into primary minima above to those formed at lower ionic strength, where secondary minima dominate the interaction, as shown in Figure 8.

**Figure 7.** The mechanical properties of a chain assembled from 3.14 \( \mu m \) PMMA particles in an aqueous solution of 0.1 M MgCl\(_2\). The chain is held with three time-shared optical traps—two stationary traps at either end and a translating trap in the center. The chain bends as the center trap is moved to the right, in good agreement with the deformation of a bending rod.
Particles interact via centrally acting forces but are free to rotate. When a compressive stress is applied, the particles in singly bonded chains easily rotate around one another until a multiply bonded structure is formed. Such particle rearrangements underlie the low-shear viscosity of weakly aggregated gels\cite{47,48} and contrast strongly aggregated chains, which exhibit an Euler instability upon compression.

**Direct Measurements of Particle Interactions**

As demonstrated above, laser tweezers are effective tools for directly measuring particle interactions associated with the mechanical properties of colloidal microstructures. Other methods developed to quantify particle pair potentials include scanning line tweezers,\cite{49,50} blinking tweezers,\cite{51} and dual traps.\cite{52} We also used laser tweezers to measure interactions on mesoscopic scales, such as the fluctuation-mediated attraction between dipolar chains that drives long-time microstructural coarsening in MR suspensions.\cite{53}

An example of a direct measurement of the interaction potential between colloids is shown in Figure 9. Two particles are held by scanning the trap rapidly along a single axis. The velocity of the trap is modulated along the line to create a weak parabolic well, biasing the particles toward the center of the line. The distribution of particle separations $d$ is given by the Boltzmann distribution,

$$P(d) = \exp\left\{\frac{-[k(d/2)^2 + U(d)]}{k_BT}\right\} \int_0^\infty dr \exp\left\{\frac{-[\kappa_1(r/2)^2 + U(r)]}{k_BT}\right\}$$  \hspace{1cm} (4)

and can be used to calculate the pair interaction $U(d)$, using the variance of the center-of-mass position of the particles $\langle R^2 \rangle = (k_BT)/(2\kappa_1)$ to find the line tweezer compliance $\kappa_1$. The weak repulsive barrier we measured in Figure 9 is consistent with an observed aggregation time scale of several minutes for a particle pair.

Line tweezer measurements are particularly useful for measuring weak interactions in complex fluids, such as those induced by polymer depletion. For instance, Yodh and
coworkers measured interactions between colloidal particles in dilute and entangled DNA solutions and suspensions of fd virus, using a method similar to those outlined above.\cite{49,50} Similar to rapid scanning, repeated positioning and releasing of colloidal particles using laser traps allows one to measure the pair interaction potential with high resolution. Crocker and Grier used these “blinking” tweezers to generate trajectories that allowed them to numerically solve the master equation for the spheres’ Markovian dynamics.\cite{51,54} Note, however, that special care must be taken when making inter-particle interactions in order to avoid nonequilibrium effects. For instance, it was found that the apparent long-range attractive interaction between two like-charged colloids near interfaces was due to hydrodynamic interactions.\cite{54,55}

\textbf{Laser Tweezer Microrheology of Polymer Networks and Gels}

Laser tweezers can also be used to measure microscopic and bulk structure and response in polymer materials. For instance, probe particle microrheology has recently emerged as a method of measuring the rheology of complex fluids. By driving the motion of embedded particles with magnetic\cite{56,57} or optical forces,\cite{58} or measuring displacement due to thermal motion,\cite{33,59-61} small sample volumes can be measured with minimal perturbation due to the extraordinarily small stresses and strains. Probe
microrheology has been of particular interest for measuring bulk and local response in polymeric systems, particularly in biological materials, such as the reconstituted cytoskeleton,\[33,61,62\] and even the rheology of individual cells.\[63,64\] Typically, one measures the thermal motion of embedded probe particles with time, known as tracer particle microrheology.\[59,60\] Particle tracking is performed using videomicroscopy,\[65\] laser tracking,\[33\] or light scattering,\[59\] depending on the length and time scales to be probed. Tracer particle microrheology relies on a generalization of the well-known Stokes–Einstein relationship for the displacement of a particle of radius $a$ in response to a force $f(\omega)$:

$$ x(\omega) = \frac{f(\omega)}{6\pi a \eta} $$

substituting the viscosity $-i\omega\eta$ with the complex shear modulus $G^*$.\[33,59,60\] The generalized Stokes–Einstein relationship (GSER) yields material responses over a wide range of time scales; however, under some conditions, the GSER breaks down\[62,66\] probably due to nanoscale fluid structure surrounding the probe, such as depletion or through enthalpically driven interactions.\[67\]

An alternative to tracking thermal motion of the probe particle is to drive particle motion with an optical trap and measure the particle response.\[58,68\] Active manipulation has been used to measure cell rheology,\[69\] the reconstituted cytoskeleton,\[70\] and membrane–cytoskeleton interactions;\[10\] however, most studies used paramagnetic particles in a field gradient to probe local response, also known as magnetic tweezers. A disadvantage of magnetic tweezers is the variation in magnetization of the paramagnetic particles, which must be known to accurately estimate the applied stress.\[28\] Laser tweezers are advantageous, because the properties that govern the optical trapping force (refractive index contrast to the medium, particle size, and shape) vary to a much smaller extent, enabling accurate calibration. The equation of motion for the bead, neglecting inertial terms, is

$$ 6\pi \eta a \dot{x} + [2(4\mu + 2\kappa) + \kappa_T]x = \kappa_T A \cos 2\pi \omega t $$

where $\eta$ is the solvent viscosity, $\kappa_T$ is the trap compliance, $\mu$ is the shear modulus, $\kappa$ is the bulk modulus, and $A$ and $\omega$ are the amplitude and angular frequency of the forcing function, respectively.\[58\] By measuring the in-phase and out-of-phase bead displacement, it is possible to measure the frequency-dependent viscous dissipation and elastic storage of the network. The particle response is $x(\omega) = D(\omega) \cos[\omega t + \delta(\omega)]$, where the amplitude is

$$ D(\omega) = \frac{\kappa_T A}{\sqrt{(\kappa_T + 2(4\mu + 2\kappa))^2 + (6\pi \eta \omega)^2}} $$

and the phase angle is

$$ \delta(\omega) = \tan^{-1} \frac{6\pi \eta \omega}{\kappa_T + 2(4\mu + 2\kappa)} $$

An example of the response is illustrated in Figure 10A for a 3 $\mu$m polystyrene (PS) particle suspended in water. The bead is oscillated using a function generator to drive
our AOD. The function generator signal was acquired in synchronization with a video frame grabber, used to capture images of the bead and perform particle tracking in real time. The measured phase angle between the laser trap and particle is small, $\delta = 4 \times 10^{-3}$ rad/s. Contrast this data to the response measured for a 10 $\mu$m PS particle in 0.1 wt% $\beta$-hairpin (D-Pro L-Pro)$_{71}$, shown in Figure 10B. The $\beta$-hairpin is a short (40 amino acid) oligopeptide that folds under basic pH conditions. The folded peptides then rapidly assemble into supramolecular structures, driven by amphiphilicity of the

![Figure 10](image_url)

**Figure 10.** (A) Response of a 3 $\mu$m PS particle (symbols) to an oscillating optical trap in water (solid line). The optical trap leads the particle by a phase angle $\delta$, given by Eq. 8. (B) Microrheology of the $\beta$-hairpin D-pro L-pro using a 10 $\mu$m probe particle. A strong elastic response is evident by the small displacements the particle translates before pulling out of the trap and the subsequent recovery. The particle jumps in on the return pass of the optical tweezer.
amino acids on opposing faces of the folded molecule. The pH sensitivity and rich microstructure make them potential materials for tissue scaffolds. Probe microrheology demonstrates the local elasticity of this dilute gel. The particle is displaced by the trap but is pulled out as the local elastic resistance increases past the maximum trapping force. A fast elastic recovery is observed before the particle is pulled back into the trap on its return (‘jump in’). From Stokes law for particle response in a purely elastic medium, we estimate that the local elastic modulus is 5 Pa.

CONCLUSIONS

This brief survey illustrates the current and future potential of optical trapping in studies of complex fluid rheology. Laser tweezers will continue to expand our ability to directly study structure–property relationships in colloids and polymers, bridging nanoscale interactions to microstructure response and bulk rheology. Several examples discussed in the case of colloids highlighted our ability to directly measure interparticle interactions, assemble micro- and mesostructures, and measure their mechanical properties. These provided insights into the mechanisms of yield and elasticity in suspensions with strong attractive interactions, such as MR fluids and particulate gels. Tweezer experiments will continue to provide new methods for testing models of suspension rheology, especially the mechanisms that underlie nonlinear response, such as strain hardening. Similarly, microrheological measurements not only enable access to extraordinarily small quantities of materials, as demonstrated by measurements of the rheological characteristics of individual cells, but also the length scales probed will be significant for studying cell–material interactions, particularly for understanding the role of substrate compliance and remodeling in tissue engineering scaffolds.

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REFERENCES


41. Iler, R.K. Relation of particle size of colloidal silica to amount of a cationic polymer required for flocculation and surface coverage. J. Colloid Interface Sci. 1971, 37, 364.


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