A-7 Light Scattering: Quasi-elastic Rayleigh Scattering Spectroscopy

From Physics 191r

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first experiment: yes

Contents

Learning Goals

- Measure the diffusion constant for microspheres in aqueous solution.
- Analyze signals using Fast Fourier Transform.
- Perform nonlinear curve fits.
- Practice laser safety.
- Understand how coherent backscattering arises from a random system.
- Observe the transition between Rayleigh Scattering and Mie Scattering.
- Study phase transitions in a binary fluid mixture.

Introduction

Light scattering has developed into a powerful tool for the study of inhomogeneities in transparent condensed matter. This includes small particles in an otherwise homogeneous medium, as well as fluctuations in the dielectric properties of material, due to changes in composition, density, or birefringence, for example. The spatial extent of fluctuations, particle size in the range of hundreds of angstroms to microns, and the temporal behavior of fluctuations, from low audio frequencies to optical frequencies can be measured. This experiment deals with fairly slow particle motion in the audio frequency range.

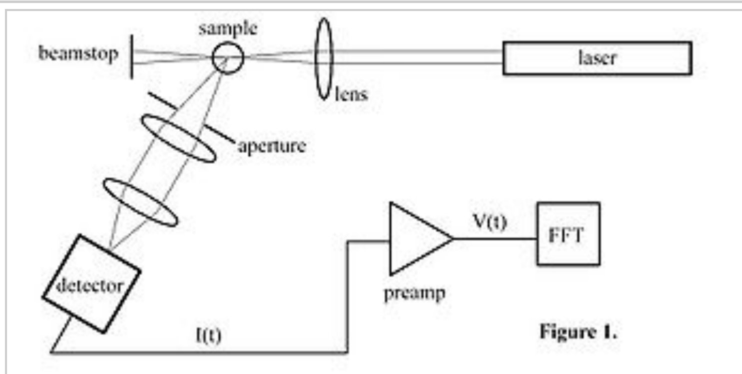
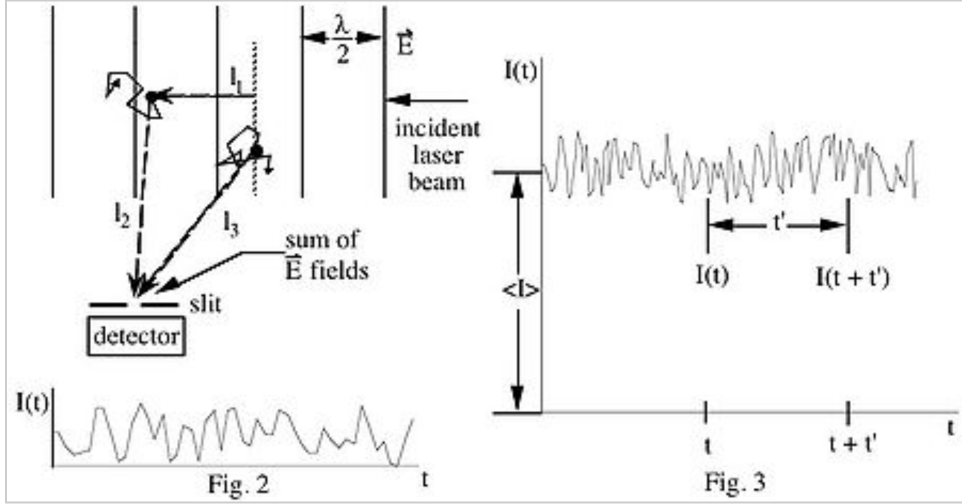


Figure 1

Dynamic light scattering is not concerned with the average intensity but rather with the time behavior of the fluctuations in the scattered intensity. A typical block diagram of the optics is shown in Figure 1.

Particles in solution continually undergo random-walk Brownian motion due to solvent collisions. This results in fluctuations in the scattered light intensity at the detector due to changes in the net interference of individual scattered waves originating from each particle. Their phases depend on the precise positions of each particle relative to both the incident exciting laser beam and the detector. This is shown in Figure 2 for just two particles in solution. In this case the scattered intensity undergoes the largest possible fluctuation, from zero – total destructive interference – to a maximum value. The mean lifetime of a fluctuation in the intensity (either a buildup or a decay) equals the average time required for random-walk diffusion to change the difference in optical path lengths to the detector ($\ell_1 + \ell_2 - \ell_3$ in Figure 2) by one-half the wavelength of light. Clearly, the larger the scattering particles, the slower the diffusion and the longer the mean decay time of the fluctuations.



Figures 2 and 3

In reality, the interference process is more complicated due to the much greater number of scattering particles. For example, if a small volume $\Delta v_i \ll \ell^3$ contains n_i particles the amplitude of the E_i-field scattered from Δv_i will be proportional to n_i. The total electric field at the detector will be the sum

$$E_{detector} \propto \sum_i n_i e^{i\Delta\Phi_i}$$

where $\Delta\Phi_i$ is the optical path length for scattering from Δv_i and *i* is summed over the illuminated sample volume viewed by the detector. The intensity is

$$I(t) \propto |E_{detector}|^2 \propto \sum_{(i,j)} n_i n_j e^{i[\Delta\Phi_i - \Delta\Phi_j]}.$$

Although $\Delta\Phi_{i,j}$ do not change with time since we are now considering scattering from a fixed volume, the amplitude of the scattering from each volume does changes with time due to diffusion of particles in and out of the volume. Hence, the photocurrent I(t) resembles the plot in Figure 3.

The mean lifetime τ of the fluctuations can be obtained by computing the autocorrelation function,

$$C(t') = \sum I(t)I(t+t').$$

This quantity is simply the product of the intensity at an arbitrary time *t* with the intensity at a later time, $t + t'$. $\sum$ indicates a running sum of such products, taken for different values of time *t*, so as to obtain a reliable statistical average of *C(t')* for a given separation time *t'*. For a uniform particle size distribution, *C(t')* is a single exponential decay,

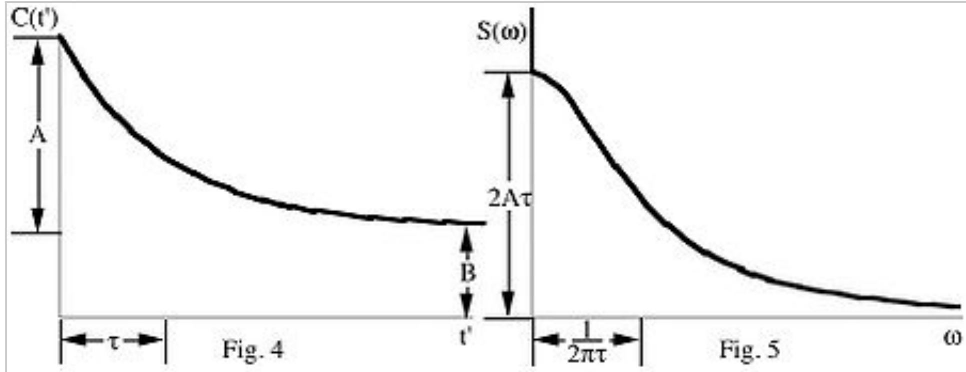
$$C(t') = Ae^{-|t'|/t} + B.$$

Baseline *B*, the value of *C(t')* at infinite time, is simply the square of the average intensity. See Figure 4.

The Wiener-Khinchin theorem (http://mathworld.wolfram.com/Wiener-KhinchinTheorem.html) states that the Fourier transform of the autocorrelation function is the power spectrum:

$$S(\omega) = \int_{-\infty}^{+\infty} C(t')e^{i\omega t'}dt'.$$

The leading term in this quantity[1] is a single Lorentzian centered on zero frequency whose width is related to the time constant τ as in figure 5. Superimposed on the Lorentzian is a delta function at zero frequency corresponding to the average intensity $\sqrt{B}$ about which the fluctuations are measured.



Figures 4 and 5

The mean decay time τ is related to the diffusion coefficient D by:

$$\frac{1}{\tau} = 2DK^2$$

where K is a geometric factor which depends on the laser wavelength λ, the solvent index of refraction and the scattering angle.

The particles' hydrodynamic radius R_h is given by the Stokes-Einstein relation[2],

$$R_h = \frac{k_B T}{6\pi\eta D}$$

where k_B is Boltzmann's constant, T is the temperature and η is the solvent viscosity.

Critical Mixing

This same formalism can be generalized to include any physical process by which the scattering amplitude from the volume element Δvi varies. For example, in any liquid or gas, molecules are continually flowing in and out of any fixed volume. For a macroscopically large ΔV_i (say 100 Å x 100 Å x 100 Å) the difference between the number flowing in or out is so small as to not be measurable. However, there are situations, particularly near "critical points" such as the temperature at which cyclohexane and methane become miscible, where the molecular interactions favor large regions that are either cyclohexane rich, or methane rich. Since cyclohexane rich regions have different scattering amplitudes than methane rich regions, the scattering from a critical solution is very much like the scattering from the solution of polystyrene balls.

The scattering amplitude of a cyclohexane/methane mixture becomes larger as temperature and concentration of the mixture get closer to the critical point. Consequently, the time constant by which the scattering amplitudes vary also becomes larger. These fluctuations can become particularly spectacular near the critical point.

Mie Scattering

(section to be written)

Coherent Backscattering

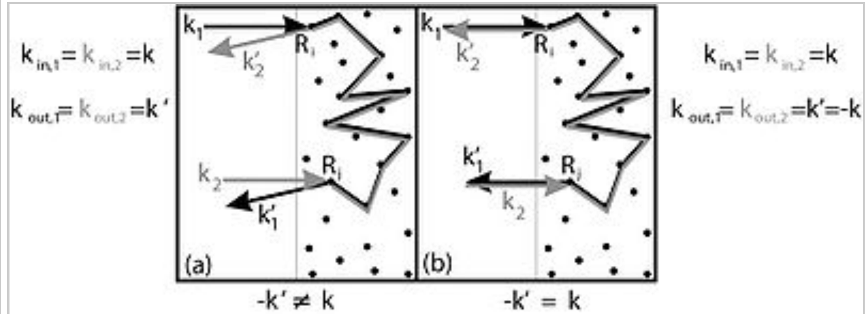


Fig. 6. Possible scattering paths for a configuration of scatterers.

Coherent backscattering is a wave phenomenon that may initially seem surprising since it arises out of a multitude of random scattering events. It manifests itself as enhanced scattering off of a semi-infinite disordered medium of scatterers in the backward direction when averaged over many configurations of scatterers. It is typically considered a signature of "weak localization", in contrast with `strong localization', in which waves inside a very strongly scattering random medium are localized to a specific region due to interference effects. The strength of localization is characterized by comparing kl with unity, where $k = \frac{2\pi}{\lambda}$, λ being the wavelength, and where l is the elastic scattering mean free path in the scattering medium. If $kl \gg 1$ then the localization is weak. If $kl \sim 1$ then the localization is strong.

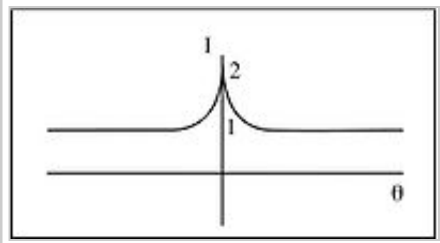


Fig. 7. Normalized intensity as a function of angle relative to 180°.

Coherent backscattering may be performed in many ways: the waves may be electrons scattering off of randomly embedded impurities in a metal, or they may be laser photons scattering inside of milk, a powder of GaAs, or polystyrene microspheres. One of the main differences between the regimes that demarcate weak and strong localization is the density of scatterers since the scattering mean free path is inversely proportional to the density. Consider coherent light incident on a configuration of scatterers such as shown in Figure 6. The scattered intensity has components caused by single scattering, uncorrelated multiple scattering, and correlated multiple scattering. The last of these gives rise to a coherent backscattering peak. Figures 6a and 6b show light rays which follow the same scattering path, but in opposite directions. They are sometimes referred to as a time-reversed pair. In Figure 6b, the outgoing rays are antiparallel to the incoming rays.

The instantaneous intensity at a distant detector as a function of scattering angle would be a random speckle pattern since the accumulated phase difference between $\mathbf{k_1}'$ and $\mathbf{k_2}'$ depends on the positions of the scatterers, Ri. However, for an average over a large number of random configurations, a peak emerges in the reverse direction as in Figure 7 since $\mathbf{k_1}'$ and $\mathbf{k_2}'$ are always in phase if $\mathbf{k}' = -\mathbf{k}$.

Apparatus

Rayleigh Scattering

Laser

As in any scattering experiment, the apparatus consists of a source of radiation, a scatterer and a detector. This experiment deals with audio frequency fluctuations in light intensity. The light source is a 5 mW He-Ne laser (Uniphase 1105P) of wavelength 632.8 nm. Never look directly at the laser beam or at the specular reflection of the beam from any surface! More than a momentary exposure can damage your retina. Wear the correct laser safety glasses whenever you align the optics or change samples. Be aware that the cylindrical glass test tube produces a significant reflection near eye level for a person seated at the computer. Block this reflection with a piece of black foamcore. Use a beam stop to absorb the light that passes through your sample without scattering. A Thorlabs optical breadboard is used to hold and position the laser, the sample, the detector and the optics.

Sample

The target is a solution of polystyrene microspheres (diameter < 500nm) in either electrolyte solution or distilled water. Old samples may be available to start with, but when you want to know the concentrations accurately, make new ones and seal them to prevent evaporation of the solvent. Take care not to waste the concentrated solution of microspheres; the cost is over $10/milliliter. Micropipets are available to measure and transfer the solution of microspheres. A typical dilution ratio is 1:5000.

Optics

Light is scattered from the region of the sample illuminated by the laser into all angles. In order to obtain a measurable signal it is necessary to collect the light scattered into a certain solid angle with a lens or lenses, and focus it on the detector. The size of the sample, the diameter of the collecting lens and the distance between them define the solid angle in which light is detected. The solid angle can also be defined by an aperture – an iris or vertical slit. The intensity fluctuations change with angle; therefore a compromise must be reached between angular acceptance and strength of signal.

Detector

The detector is an EG&G Judson YAg-100A photodiode. This device has an active area of 5.1 mm^2 and noise-equivalent power of 1.2 x 10^{-13} W/Hz at 1000 nm, i.e., the mean square fluctuations in the photodiode current in the dark are equivalent to illumination by 1.2 x 10^{-13} W. When struck by a photon of sufficient energy, an electron-hole pair is created in the photodiode. Proper biasing of the diode generates a current from this event. Therefore the current through the diode will be proportional to the light intensity by which the diode is illuminated. The diode housing holds an interference filter tuned to the color of the laser light.

Amplifiers

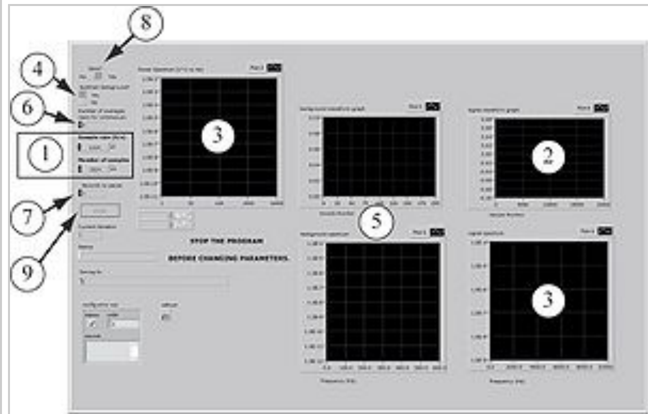
The photocurrent is converted to a voltage by a transimpedance preamplifier (EG&G PA-6-60 or PA-7-70). Choose from the two models available; each has three settings, which provide different gains and bandwidths. A typical gain is 1 V/μA. The voltage output from the preamplifier is fed to a two-channel 24 dB/octave filter with amplifiers (Ithaco 4302 or PAR 113), which may be used as a low pass filter to remove effects of aliasing and/or a high pass filter to eliminate 60-cycle noise and/or a secondary amplifier.

Data collection

Data acquisition and processing are performed by a computer and a National Instruments analog to digital converter card (PCI-6034E). A LabVIEW program called "fft_mx.vi", which acquires a time-domain signal from the amplifier, controls the PCI card. See the figure below for more information. The user determines sample rate and number of samples. The time domain signal is Fourier transformed to yield the power spectrum.

You can practice using the program by analyzing simple waveforms from a function generator (Wavetek 182A). The input to the FFT is the “AI0” BNC jack of the NI BNC-2110 interface box. Choose floating or grounded source with the “FS/GS” switch as appropriate.

The fft program functions as both an oscilloscope and as a spectrum analyzer. Investigate and understand the effects of “SAMPLE RATE” and “NUMBER OF SAMPLES” on the frequency-domain signal. Data may be saved to a text file. This file format cannot be read back into fft_mx.vi, but it may be viewed with “texttograph.vi”, or any graphing software. The averaging feature of the program is extremely useful for averaging out time-varying frequency components, but it will not increase your signal to noise ratio. Therefore take precautions to minimize background signal from room light and extraneous scattered laser light.



LabVIEW VI for Rayleigh Scattering

- (1) SAMPLE RATE and NUMBER OF SAMPLES are important parameters. They determine the bandwidth and resolution of the fft spectrum. Understand this, and choose them appropriately before starting systematic measurements on microsphere samples.
- (2) Graph # 2 displays the time domain signal.
- (3) Graphs # 3 display the power spectrum.
- (4) The program has an automatic background subtraction feature. It may be more instructive to measure and save the background signal and subtract it during analysis if it is significant.
- (5) Indicators # 5 are the time domain signal and power spectrum of the background.
- (6) Control # 6 sets the NUMBER OF AVERAGES. The program will acquire this number of spectra, and compute the numerical average. One means to acquire a single spectrum then stop. Zero means to acquire single spectra continuously.
- (7) Control SECONDS TO PAUSE delays the start of execution. This is useful for leaving the room (without flooding the detector with ambient light) during long acquisition periods.
- (8) The SAVE? switch MUST be switched to Yes BEFORE running the vi if you want to save data.
- (9) Use this STOP button to end execution of the program.

Critical Mixing

(section to be written)

Mie Scattering

(section to be written)

Coherent Backscattering

Laser

The light source for coherent backscattering is a laser diode (Hitachi HL6501MG). The maximum optical power is 30 mW at 657 nm. See the bench notes for a plot of laser power vs. current. This is a class IIIb laser. It can damage your vision. Wear the correct laser safety glasses whenever you align optics. Use an IR card to view the beam location. The card converts the red light to orange, which is visible through the glasses. Always insert the card into the beam in such a way that the specular reflection from the card goes downward.

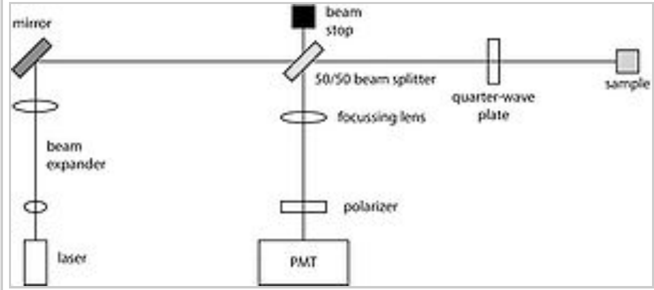


Figure 8. Coherent backscattering schematic.

The laser is housed in a temperature-controlled mount (Thorlabs TCLDM9). The Thorlabs ITC510 supplies current to the laser diode and regulates the temperature. When you are ready to turn the laser on, ask the faculty or staff for the key. The default temperature in units of resistance is 11.75 kΩ, a little below room temperature. Turn on the temperature control with the left-hand pushbutton marked ADJUST:ON. The controller monitors the resistance of a 10 kΩ thermistor, and supplies current in either polarity to thermoelectric coolers to cool or heat the laser as needed. Several minutes are needed for the temperature to stabilize. Next check that the laser current control knob is fully CCW. Then turn on the current with the right-hand pushbutton marked ADJUST:ON. Slowly turn up the current above the lasing threshold (to approx. 48 mA). For alignment, just a few mW are enough. When you are ready to take backscattering data, you can turn the current up to maximum (80 mA).

Optics

Referring to Figure 8, note that the first optic in the beampath is a beam expander. Increasing the size of the beam exposes more of the sample to photons. In order to study the light scattered in the backward direction, we use a 50/50 beamsplitter. Half of the incident beam goes to a beam stop, while the other half irradiates the sample. Of the scattered light that reaches the beamsplitter, again half is lost while half goes to the detector. A quarter wave plate at 45° to the incoming polarization, in conjunction with a polarizing filter selects the scattering events that preserve helicity. A 500 mm lens focuses the light onto the detector.

Detector

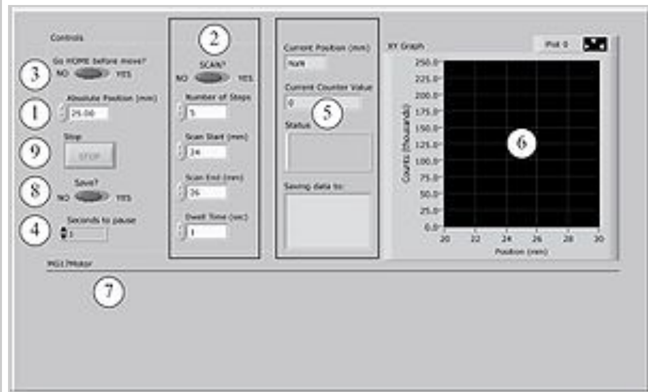
The detector is a Hamamatsu H6780-20 photomultiplier tube assembly (PMT). It has an integrated high voltage supply; the power input is 12 V_{dc} from the Stancor power adapter. A separate green wire connected to the optical bench grounds the aluminum chassis in which the PMT is mounted. Output pulses are available at the rear-panel BNC connector, and a potentiometer mounted on top of the aluminum chassis controls the gain. A typical gain is 10^5. A 100-micron pinhole is fixed over the entrance aperture of the PMT. The PMT must never be exposed to room lights, even when it is turned off. Keep the black plastic envelope over the PMT except when it is actively in use.

Electronics

The output pulses from the PMT module are current pulses, so care must be taken to terminate them properly. A discriminator/amplifier (Mechtronics 511) converts the current pulses to voltage pulses and amplifies them. It rejects pulses smaller than a threshold, which is set by a recessed potentiometer on the front panel. Choose a reasonable gain and threshold. Positive output pulses from the discriminator can be counted by the National Instruments card (the same PCI-6034E used for Rayleigh scattering), or the Tennelec ratemeter (TC 596). The analog display of the ratemeter makes it easy to find a peak in counts, so use this when fine-tuning the angular position of the beamsplitter as described below.

Data Collection

A translation stage (Thorlabs LNR50) is used to scan the PMT across the optical axis. A LabVIEW program called cbs.vi controls the position of the stage and counts pulses. This is a LabVIEW container for commercial software that controls the translation stage, with an additional interface to the counter of the PCI-6034E. The input to the counter is the “USER1,” BNC jack of the National Instruments BNC-2110, which in turn is connected by a purple wire to “PFI8.” The program can either move the stage to a specific position and stop, or step through a range of positions, counting for a fixed time at each step. A stepper motor moves the stage; the range of motion is 50 mm. A Thorlabs BSC101 controller powers the stepper motor model DRV014. On the first run after turning on the power, send the stage to its "home" position, one extreme of travel defined by a limit switch. This establishes the zero location. The center of travel is then at 25 mm.


LabVIEW VI for Coherent Backscattering

- (1) The simplest mode of cbs.vi is to send the stage to an ABSOLUTE POSITION defined by this control.
- (2) If control SCAN? is set to No, cbs.vi send to stage to ABSOLUTE POSITION, then stops. If SCAN? is set to Yes, the four controls determine the scan parameters. The stage moves to SCAN START, counts pulses there for DWELL TIME seconds, then moves toward SCAN END in NUMBER OF STEPS intervals, counting at each intermediate position.
- (3) Control # 3 should be set to Yes on the first run after powering on the stepper motor controller. This sets the zero of position.
- (4) Control SECONDS TO PAUSE delays the start of execution. This is useful for leaving the room (without flooding the detector with ambient light) during long acquisition periods.
- (5) Indicators # 5 give information about the scan or motion as they are in progress.
- (6) Graph # 6 shows count rate as a function of position.
- (7) Indicator # 7, MG17MOTOR, is the commercial motor control program.
- (8) The SAVE? switch MUST be switched to Yes BEFORE running the vi if you want to save data.
- (9) Use this STOP button to end execution of the program.

Alignment

A few hints about alignment will help to locate the backscattering cone. Keep the PMT covered until the end. Set the laser current at about 48 mA. This gives you a few mW of optical power. If aligning from scratch, remove all optics except the steering mirror and sample. Center the beam on the sample with the mirror and check that the beam is parallel to the optics table at the correct height, and that it follows rows of tapped holes in the table. Put in the beam expander lenses (focal lengths 100 mm and 400 mm) one at a time. Adjust the position and height to recover the beam path. Separate the lenses by a distance equal to the sum of their focal lengths. Place the quarter waveplate just upstream of the sample. Rotate it and the sample cell slightly, so that the reflections from their surfaces go off-axis.

Next insert the beamsplitter on the goniometer mount at about 45° to the optical axis. Put a mirror in front of the sample and adjust it so that the beam reflects back on itself. Use an index card with a hole punched in it. Now a partial reflection from the beamsplitter goes toward the detector. Place an iris, open a few mm, in front of the PMT, approximately centered on the translation stage to act as a temporary target. Rotate the beamsplitter mounts in both theta (horizontal) and phi (vertical) until the beam is centered on the iris. Insert the 500 mm focusing lens and adjust it so that the beam hits the iris again. Locate it 500 mm from the detector. Insert the polarizing filter with its transmission axis parallel to the incident polarization. Open the iris, check that the translation stage is in the center of its travel and turn off the room lights. Insert a neutral density filter (ND 3.0). Attenuate the light further by rotating the polarizing filter until you almost lose sight of the focused laser spot (without the laser glasses). Remove the black cover from the PMT, and repeat the angle adjustments to center the dim spot on the actual pinhole. Plug in the PMT; look at the pulses directly with the oscilloscope. Use the amp/discriminator to reject noise. The discriminator output can drive the ratemeter. Use the ratemeter to optimize theta and phi. Set the scale to a reasonable value, 10 k/sec to 100 k/sec. Use the polarizer as a variable attenuator to keep the reading on scale as you center the spot on the pinhole. Remove the alignment mirror.

Sample

Start with a sample of stock 10% microsphere solution. One-micron spheres are typical; 0.4-micron spheres have been shown to work well also. Set the quarter wave plate and polarizing filter to suppress incoherent scattering modes. These modes produce a very broad maximum whereas the coherent scattering is found within only a few milliradians of the backward direction. Check that all stray reflections of laser light are confined within the footprint of the bench. Turn up the laser power, and make a scan. You may need to re-optimize phi with the ratemeter, this time using the CBS signal itself.

EXPERIMENTAL PROCEDURE

Required

- Measure the frequency spectrum as a function of a) angle b) size of scatterers, and c) concentration of scatterers. Calculate the diffusion constant from your data and compare it with the expectation based on the Stokes-Einstein relation. Alternatively, assume that the Stokes-Einstein result is correct and either calculate the radius of the microspheres or use the nominal radius supplied by the manufacturer to calculate the value of a physical constant such as Boltzmann’s constant.
- Measure the coherent backscattering cone for samples with different scattering lengths. See the paper by Corey (1995).

Optional

- Study Rayleigh scattering as a function of incident light polarization.
- Study critical mixing in a binary fluid mixture (methylcyclohexane / perfluoromethylcyclohexane).
- Measure cross sections for Rayleigh and Mie scattering for different size microspheres as a function of optical wavelength. See the paper by Cox (2002).

References and Notes

1. ↑ See Clark *et. al*., Am. J. Phys. **38**, 575 (1970), for example.
2. ↑ See Morse, *Thermal Physics*, 232ff, for example

Additional Reading

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Mie Scattering

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Multiple Scattering

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Bench Notes

Rayleigh Scattering

- National Instruments PCI 6034E multifunction I/O card (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/6034e.pdf)
- Interference Filter (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/interferencefilter.pdf)
- PAR 113 Preamplifier (http://www.fas.harvard.edu/~phys191r/Bench_Notes/E1/Preamp.pdf)

- Ithaco Low-Noise Voltage Preamplifier (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/ithacopreamp.pdf)
- Ithaco 4302 Dual Filter (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/ithaco4302.pdf)
- Microspheres (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/Microspheres.pdf)
- Uniphase He-Ne Laser (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/uniphase.pdf)
- Wavetek 182A Function Generator (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/Wavetek182.pdf)

Coherent Backscattering

- Hamamatsu H6780-20 PMT (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/h6780_20_data.pdf)
- Hamamatsu H6780-20 test sheet (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/h6780_20_test.pdf)
- Translation Stage (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/lnr50.pdf)
- Stepper Motor Controller (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/bsc101.pdf)
- Laser Diode (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/hl6501mg.pdf)
- Laser Diode -- Schematic (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/HL6501MG_schematic.pdf)
- Laser Diode Threshold Plot (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/laser_threshold_plot.pdf)
- Laser Diode Mount (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/tcldm9.pdf)
- Laser Diode Mount -- Schematic (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/TCLDM9_schematic.pdf)
- Collimating Lens (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/C230TME.pdf)
- Laser Diode Controller (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/itc510.pdf)
- MechTronics 511 Amp/Discriminator (http://www.fas.harvard.edu/~phys191r/Bench_Notes/A7/mechtronics511.pdf)

Appendix: Speckle Patterns

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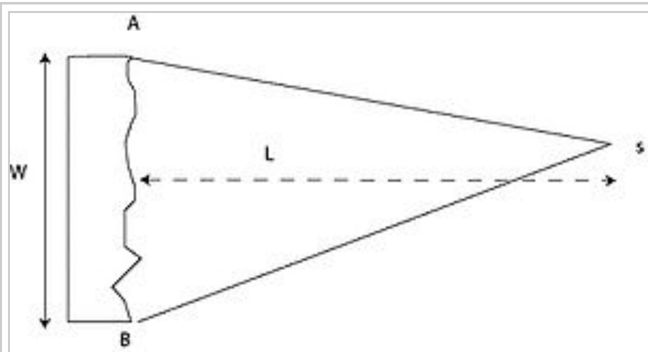
The figure at right illustrates the schematics for speckle formation due to scattering from a rough surface $\overline{AB}$. Assuming that there is a bright spot at point "S" on a screen. This bright spot occurs because the statistical distribution of the phases from all of the scattering from all of the different points on the surface happen to have phase relations for which the constructive interference dominates. Note that the path lengths $\overline{AS}$ and $\overline{BS}$ are



Schematic for speckle formation.

$$r_{AS} \approx L + \frac{1}{2}\frac{(s-W/2)^2}{L} \approx L + \frac{1}{8}\frac{W^2}{L} - \frac{1}{2}\frac{sW}{L}$$

$$r_{BS} \approx L + \frac{1}{2}\frac{(s+W/2)^2}{L} \approx L + \frac{1}{8}\frac{W^2}{L} + \frac{1}{2}\frac{sW}{L}$$

Although the path difference is

$$\delta r_{AB} \approx \frac{sW}{L}$$

the phase of the wavefronts at A and B could have been such that the waves interfered constructively at S. The point is that there are some distribution of waves emanating from the region between A and B for which there are more waves that interfere constructively at S than not. The issue now is that if S changes by δ*s* the these phases will vary; however, if

$$\Delta\left[\delta r_{AB}\right] = \frac{W}{L}\delta s \ll \frac{\lambda}{2}$$

the various phases can not vary by enough to make a significant difference in the constructive interference.

The conclusion is that the speckle width is of the order of

$$\Delta s_{speckle} \sim \frac{\lambda L}{2W}$$

As a practical matter the width W is either (a) the size of the sample, or (b) the coherence length of the illuminating laser.

Although the amplitude of $\langle I(t,\vec{r})I(t+\tau,\vec{r})\rangle$ or $\langle I(t,\vec{r})I(t+\tau,\vec{r}+\delta\vec{r})\rangle$ are different I don't think that the time dependence (i.e. spectra) of the diffuse scattering is different for these two.