Measurements of the Rayleigh Ratio of Some Pure Liquids at Several Laser Light Wavelengths

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Measurements of the Rayleigh ratio of some pure liquids at several laser light wavelengths

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The Rayleigh ratios $R_{v,v}$ for vertically polarized incident and scattered radiation of benzene, toluene, carbon tetrachloride, and cyclohexane have been measured at a scattering angle of 90° and at 25 °C using argon–ion and krypton–ion lasers at wavelengths 488, 514.5, and 647.1 nm. The problem of the change of the scattering geometry with the refractive index of the investigated liquid was solved by a suitable experimental procedure. Errors are estimated to be in the range of 1%–2%.

I. INTRODUCTION

The particular coherence properties of laser radiation has led in the last 20 years to new kinds of light scattering experiments not feasible with traditional light sources. These dynamic light scattering experiments have found an increasing number of applications in physics, chemistry, biology, and medicine.[1]

On the other hand, the high energy density, small beam divergence, and the high pointing and intensity stability of some laser beams has encouraged the use of laser sources in the traditional static light scattering experiments, where the time averaged scattered intensities are studied.[2-5] Now both dynamic and static light scattering experiments can be performed with the same laser source and spectrometer.[2-6]

One of the main applications of static light scattering experiments, i.e., the determination of molecular weights of macromolecules, requires absolute intensity measurements. Consequently there is an increasing demand for standards with precisely known absolute scattering power for the calibration of the light scattering photometers at laser light wavelengths.

In the older experiments, using mainly the mercury arc as a light source, the scattering by pure liquids, such as benzene, has very often been used as a primary calibration standard. Therefore, many efforts have been spent for the determination of the Rayleigh ratio of benzene and other liquids especially at the 436 and 546 nm lines from the mercury spectrum. The initially rather widely different results, have been critically reviewed by Kratohvil et al.[7] and Utiyama[8] who provide "best" values for calibration purposes.

At laser wavelengths however there are only a few measurements of the Rayleigh ratio of pure liquids,[9-14] most of them at the 633 nm line of the He–Ne laser. The present knowledge of the Rayleigh ratio of benzene at the most frequently used laser line wavelengths in light scattering, i.e., 633 nm for the He–Ne lasers and 488 and 514 nm for the argon ion lasers, can be summarized as follows:

(1) Within a few percent there is an agreement between the different experimental values at 633 nm.[9,11,14] These values agree also with Kratohvil best values at 436 and 546 nm extrapolated to 633 nm using an equation for the wavelength dependence of the Rayleigh ratio.[11]

(2) For the argon–ion laser lines only two experimental

values are reported at 488 nm.[12-13] There is a difference of about 15% between these two values.

In this contribution we report some measurements of the Rayleigh ratio of some pure liquids at several Ar–ion and Kr–ion laser line wavelengths. Since most of these lasers provide linearly polarized incident radiation and since most detection systems show variations in the sensitivity for different polarizations, Rayleigh ratio $R_{v,v}$ for vertical polarized incident and scattered radiation are determined.

The problem of the change of the scattering geometry with the refractive index of the scattering sample was solved by an experimental procedure.

II. PRINCIPLES OF THE CALIBRATION PROCEDURE

The radiation of most lasers used in light scattering experiments is linear polarized, usually with the electrical field vector perpendicular to the incident and scattered directions (vertical polarization). As a consequence, Rayleigh ratio R_v for vertically polarized incident radiation are preferable to the Rayleigh ratio R_u for unpolarized incident radiation (Hg sources). There are essentially two possibilities for the detection of the scattered radiation. In the first no analyzer is placed before the detector. If the sensitivity of the detection system does not show variations to different polarizations of scattered light, or if corrections for the variation of the sensitivity are taken into account, this configuration will yield the Rayleigh ratio R_v. In the second configuration an analyzer is placed in front of the detector. If the polarization defined by the analyzer is parallel with the polarization of the incident radiation the Rayleigh ratio $R_{v,v}$ will be obtained.

Since most detection systems show variations in the sensitivity to different polarizations the Rayleigh ratio $R_{v,v}$ is the most suitable for calibration purposes.

For simple fluids the relation between the three mentioned Rayleigh ratios at a scattering angle of 90° is given by[8]

$$R_u = \tfrac{1}{2}R_v(1+\rho_u) = \tfrac{1}{2}R_{v,v}(1+3\rho_v), \tag{1}$$

where ρ_u and ρ_v are the depolarization ratios for unpolarized and vertical polarized incident radiation, respectively. The relation between ρ_u and ρ_v is for simple liquids given by

$$\rho_u = 2/(1+\rho_v^{-1}). \tag{2}$$

The Rayleigh ratio or factor $R_{v,v}$ can be defined as the ra-

diant power $P_{v,v}$, scattered per unit solid angle σ, per unit irradiance I_0 of the illuminating beam, per unit scattering volume V[9]:

$$R_{v,v} = \frac{P_{v,v}}{I_0 V \sigma}.$$ (3)

The scattered volume in Eq. (3) is the volume of the illuminated sample from which the scattered radiation is observed.

Equation (3) can be redefined in terms of the radiant power P_0 of the illuminating beam[9]:

$$R_{v,v} = \frac{P_{v,v}}{\sigma V} \frac{A_0}{P_0} = P_{v,v} \frac{1}{\sigma 1 P_0},$$ (4)

where A_0 is the area illuminated and $1 = V/A_0$ is a length viewed by the detector parallel to the incident beam.

In an experiment the total radiant power $P_{v,v}$ falling on the photomultiplier cathode is measured in arbitrary units.

Hence the calibration procedure of a spectrometer consists in the determination of the calibration factor $1/\sigma 1$ or $1/\sigma 1 P_0$ if incident power is kept constant during the experiment.

Although in principle the factor $1/\sigma 1$ can be evaluated from the geometrical and optical arrangement of the spectrometer, it turns out that in practice these evaluations are not feasible with sufficient accuracy[9]. So indirect determinations of the calibration factor are preferable.

In the past methods were used based on the relation between the energy loss by the scattering process and the attenuation of the incident beam by the (scattering) specimen.[7,8] For small isotropic Rayleigh scatterers this relation can be written as

$$\tau = \frac{8\pi}{3} R_{v,v}.$$ (5)

In Eq. (5) the turbidity τ is the (absolute) measure for the energy loss by attenuation and $R_{v,v}$ is the measure of the energy loss by scattering. Combining Eqs. (4) and (5) yields the following for the calibration factor:

$$\frac{1}{\sigma 1 P_0} = \frac{3}{8\pi} \frac{\tau}{P_{v,v}}$$ (6a)

or

$$\frac{1}{\sigma 1} = \frac{3}{8\pi} \frac{\tau}{P_{v,v}/P_0}.$$ (6b)

So combined measurements of the turbidity and the scattered power of one particular specimen yield in principle the calibration factor. Since for pure liquids the turbidity cannot be measured with a conventional spectrophotometer, calibration experiments are often performed with colloidal dispersions of silica in an aqueous medium (Ludox, Siton X, ...). There are, however, two practical problems associated with the use of Ludox dispersions. First the ratio $\tau/P_{v,v}$ is not constant as a function of the colloid concentration. This effect is mainly attributed to attenuation of the incident and the scattered radiation, to multiple scattering by the specimen, and to interactions between the dispersed particles.[15–17] Since all these effects vanish in the limit of zero colloid concentration, this drawback can be overcome easily by determinations of the ratio $\tau/P_{v,v}$ as a function of the colloid concentration and extrapolations to infinite dilution.

In this procedure the measured turbidity or scattered power can be used as a measure of the concentration.

A second problem arises from the fact that the scattering volume V and the illuminated area appearing in the calibration factor is dependent on the refractive index of the specimen. As a consequence the Rayleigh ratio (i.e., the absolute measure) of an unknown scatterer can only be calculated using Eq. (4) if the calibration factor has been determined with calibration samples with the same refractive index of the unknown specimen, unless the dependence of the scattering volume, illuminated area and of the scattered power on the specimens refractive index is known. The latter, often called refractive index correction, can in principle be evaluated from the optical and geometrical parameters of the spectrometer.[18]

However, these calculations have led in the past to different results. Moreover most manufacturers of commerical laser–light spectrometers usually do not provide sufficient information allowing to derive the exact relation for the refractive index correction.

In order to circumvent this problem, the dependence of the calibration factor on the sample refractive index was assessed experimentally by determinations of the calibration factor with colloidal dispersions of silica in aqueous media ($n \sim 1.33$) and in toluene ($n \sim 1.50$).

III. EXPERIMENTAL

A. Equipment

The light sources were a Lexel 85.5 Ar–ion laser, a Coherent Innova 90-3 Ar–ion laser and a Coherent 52-K krypton ion laser retrofitted with an Innova 90-K plasma tube.

All these lasers were operated in the light control mode. Incident power variations were typically of the order of $\pm 0.2\%$.

The light scattering spectrometers were two Malvern 4300 systems, including EMI 9863 KB100 phototubes and a K 7023 correlator counter. The output of this instrument was fed into a HP 9825 A calculator for data analysis.

At the wavelengths of 488 and 514.5 nm, interference filters were placed in front of the photomultiplier.

B. Materials

The pure liquids were Aldrich and Mercckx products "pro analysis." Aqueous silica suspensions were obtained by diluting a Ludox HS 40 solution (Dupont de Nemours Co.) at a pH of about 10. Dispersions of stearyl silica in toluene[19] were obtained graciously from Professor Vrij (Van't Hoff laboratoria, State University of Utrecht, The Netherlands).

All samples were cleared from dust by passing the aqueous specimens through Sartorius SM11107 filters (pore sizes 0.2 μm) and the organic liquids and dispersions through Sartorius SM11807 filters (pore size 0.2 μm). The colloid concentration of the dispersion varied between about 0.4 to 2.5 wt. %.

C. Scattered power measurements

All measurements were performed at 25.0 $\pm$ 0.1 °C. The

incident power was adjusted to about 10 mW. For the coherent Ar– and Kr–ion lasers this was achieved by placing neutral density filters in the incident beam.

Photon counting techniques similar to those described by Pike *et al.*[11] were used. Typical count rates of scattered radiation were of the order of 10^3 to 10^5 counts s^{-1}. Dark current and background count rates were of the order of 5–10 counts/s. The dead time of the detection system (55 ns) was determined using the procedure of Johnson *et al.*[20]

All measurements were corrected for dark current, background light, and dead time effects. For the colloidal dispersions the excess scattered power was determined as the difference in count rate between the dispersion and the dispersing liquid.

For each sample a series of at least ten measurements, each during 10 s, was recorded. The fractional error on the measurements was estimated in a way similar as done by Pike *et al.*[11] In all cases it was less than 1 part in 1000.

D. Turbidity measurements

The difference in absorption between the dispersions and the solvents was measured with a Kontron Uvikon 810 double beam spectrophotometer, using 10 cm path length cells (Hellma). The bandwidth was 2 nm and the temperature of 25.0 ± 0.1 °C was controlled by circulating water from a Neslab model EX-200 thermostat through a jacket surrounding the cells. The measurements were carried out under control of an Apple IIe microcomputer. Data analysis was also performed with this computer.

In order to verify the linear dependence of the absorption with cell length, the absorption of a suspension was measured with cells with path lengths of 1, 2, 5, and 10 cm. Within better than the experimental accuracy, actually within ± 0.001 absorption unit, the absorption was found to be direct proportional to the cell length.

Errors on the measured turbidity may arise from the finite angle of acceptance of the spectrophotometer used for the turbidity measurements.[21,22] In the Rayleigh–Gans–Debye approach, the turbidity τ_α as determined at a finite acceptance angle α, defined as the sum of half the angle of divergence or convergence of the incident beam and half the angle subtended by the detector, is related to the turbidity τ for an infinite small acceptance angle by

$$\tau_\alpha = \tau(1 - C_\alpha)$$

with

$$C_\alpha = \frac{3}{8} \int_0^\alpha P(\theta)(1 + \cos^2\theta)\sin\theta\, d\theta,$$

where $P(\theta)$ is the particle form factor.

Using $P(\theta) \cong 1$ for the investigated particles, the correction factor C_α becomes

$$C_\alpha = \tfrac{3}{4}(1 - \cos\alpha).$$

From the optical diagram of the used spectrophotometer we estimated $\alpha \cong 6°$, so that $C_\alpha \cong 0.004$.

The effect of the acceptance angle was investigated experimentally by measurements of the absorption of a Ludox dispersion in a 10 cm cell. Three acceptance angles were investigated: two by placing diaphragm's with diameters, re-

TABLE I. Turbidities τ in m^{-1} and ratios turbidities to scattered power $\tau/P_{u,v}$ (10^{-6}m^{-1}) of Ludox and stearyl silica dispersions at 514.5 nm wavelength and 25 °C.

Ludox dispersion		Stearyl dispersion	
τ	$\tau/p_{v,v}$	τ	$\tau/P_{u,v}$
1.46_0	7.85_5	2.66_9	9.69_2
1.84_0	7.73_8	3.00_3	9.85_4
2.54_7	8.16_8	3.31_6	9.83_7
2.98_2	8.30_4	3.68_0	9.99_0
3.44_8	8.43_8	4.12_5	10.19
3.71_5	8.56_5	4.47_0	10.21
4.11_1	8.67_9	5.45_8	11.36
4.52_8	8.70_4	5.98_6	11.37
4.79_0	8.88_5	5.99_7	10.65
5.02_5	8.89_2	6.35_2	10.76

spectively, 1 and 3 mm before and behind the cells; for the third angle no diaphragm was used. Within the experimental accuracy (± 0.001 absorption unit), no difference in absorption was observed.

The error of the absorption due to the finite bandwidth of 2 nm was estimated according to Walstra[21] [his Eq. (6)]. In the worst case the error was about 0.02%.

The probable errors on the turbidity are about $\pm 2.10^{-2}$ m^{-1}.

E. Particle size measurements

The calibration of the spectrometer based on the combination of turbidity and intensity measurements [Eqs. (5) and (6)] requires, in principle, scatterers of linear dimensions small compared to the wavelength of light. Therefore, the size of dispersed silica particles was determined by small angle x-ray scattering (SAXS). The experimental equipment consisted of a Siemens Kristalloflex 805 HV power supply, a Siemens x-ray tube type FK 60-20 with copper anode, a Kratky compact camera (Anton Paar), and an Inel Leti x-ray position sensitive proportional counter.

From the slope of $\ln I(h)$ vs h^2, $I(h)$ being the scattered intensity and $h = 4\pi \sin\theta/\lambda$, where λ is the x-ray wavelength (1.54 Å) and 2θ the scattering angle, the radius of gyration R_G^2 of the particles was determined. Assuming a spherical shape the diameter was obtained from

$$d = 2\sqrt{\tfrac{5}{3}R_G^2}$$

yielding the following diameters: ludox particles 15.6 ± 0.2 and 18.8 ± 0.2 nm for the stearyl silica. Since for the worst case the diameter is about 25 times smaller than the shortest wavelength of the light, no measurable effect of particle size is to be expected.

IV. RESULTS AND DISCUSSION

A. Calibration

In Table I and Fig. 1 typical results of scattered power and turbidity measurements with Ludox and oleophilic silica at 514.5 nm are presented.

Clearly the ratio $\tau/P_{v,v}$ is dependent on the turbidity of the sample. As mentioned this effect is usually attributed to
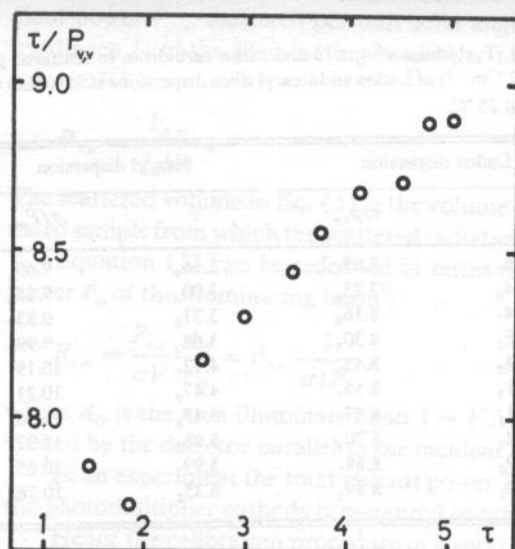
FIG. 1. Ratio turbidity to scattered power $\tau/P_{v,v}$ (10^{-6} m^{-1}) vs turbidity (m^{-1}) for aqueous silica dispersions at 514.5 nm wavelength and 25 °C.

the attenuation of primary and scattered radiation, to multiple scattering by the specimen, and to interactions between the dispersed particles.

Since the effect of multiple scattering becomes only detectable at turbidities above 10 m^{-1},[23] we exclude this effect for our measurements.

The effect of interactions can be estimated as follows.

In the case of interactions the expressions for the scattered intensities and turbidity are modified as follows:

$$P_{v,v} = P_{v,v}^0 S(q),$$

$$\tau = \tau^0 \frac{3}{8} \int_0^\pi S(q)(1 + \cos^2 \theta) \sin \theta \, d\theta,$$

where $P_{v,v}^0$ and τ^0 represent the power and turbidity that would be observed without interactions and $S(q)$ is the static structure factor; q is given by

$$q = 4\pi n \sin(\theta/2)/\lambda_0.$$

Here θ is the scattering angle, n the solvent refractive index, and λ_0 the wavelength *in vacuo* of the incident radiation.

The structure factor can be evaluated for a hard-sphere-type interaction[25] yielding

$$S(q) = 1 - \frac{4\pi\rho}{q^3}(\sin q\sigma - q\sigma \cos q\sigma).$$

Here σ is an effective hard-sphere radius. This gives the following expression for the ratio:

$$\tau/P_{v,v} = \frac{\tau^0}{P_{v,v}^0} Q$$

with

$$Q = \left(\frac{3}{8} \int_0^\pi S(q)(1 + \cos^2 \theta) \sin \theta \, d\theta \right) / S(\theta = 90°).$$

Assuming $\sigma = 1$ to 1.5 times the particle diameter d, the numerical value of Q ranges between 0.9995 and 0.9999 so that the effect of interactions may be neglected.

The effect of attenuation of primary and scattered beam was treated following Maron and Lou,[15] yielding

Wavelength (nm)	Calibration factor		Ratio of calibration factors	Ratio n^2
	$n = 1.33$	$n = 1.50$		
488	4.70 ± 0.09	5.8 ± 0.1	1.23 ± 0.03	1.27
514.5	8.83 ± 0.09	10.93 ± 0.08	1.24 ± 0.03	1.27
647.1	5.2 ± 0.1	6.69 ± 0.05	1.29 ± 0.03	1.25

$$\ln \frac{\tau}{P_{v,v}} = \ln(\tau/P_{v,v})_0 + l_1 \tau,$$

where $(\tau/P_{v,v})_0$ represents the ratio value without attenuation and l_1 is an optical path length (in principle, the path length through the specimen cell). Clearly whatever the cause of the dependence of the ratio $\tau/P_{v,v}$ on the colloid concentration, the effect vanishes in the limit of zero colloid concentration.

So the calibration factors are calculated using the values of $\tau/P_{v,v}$ extrapolated to zero excess turbidity. Table II summarizes the values of the calibration factors at different wavelength, together with solvent refractive index.

Since at each wavelength the incident power P_0 was kept constant, the differences in calibration factor as obtained with the Ludox samples on the one hand and with the oleophilic silica on the other hand, reflects directly the variations of the scattering geometry with the refractive index of the specimen. The ratio of the two calibration factors obtained at each wavelength was found within the experimental accuracy to be equal to the square of the ratio of the refractive indices of the dispersion media (see Table II).

B. Rayleigh ratio of pure liquids

The scattered power of several liquids were also determined during the calibration procedure, i.e., they were measured with the same incident power P_0 as for the determination in the calibration factor. The Rayleigh ratios listed in Table III were calculated according to

$$R_{v,v} = P_{v,v} \frac{1}{\sigma 1 P_0} \left(\frac{n}{n_{ref}} \right)^2.$$

Here n is the refractive index of the investigated liquid and n_{ref} is the refractive index of the dispersion liquid used for the

TABLE III. Rayleigh ratio $R_{v,v}$ (in 10^{-4} m^{-1}) at 25 °C and 90° scattering angles for several liquids.

Wavelength (nm)	Benzene	Toluene	Carbon tetrachloride	Cyclohexane
This work				
488	29.1	31.0	17.1	15.3
514.5	23.0	24.2	13.5	11.7
647.1	7.81	8.50	4.97	4.27
Finnigan and Jacobs (Ref. 12)				
488	32.2	29.7	19.7	
Bender et al. (Ref. 13)				
488	27.5	29.5	16.8	14.9

TABLE IV. Ratio of isothermal compressibility to the square of the isobaric expansivity β/α^2 in 10^{-4} m³ °C² J⁻¹ at 25 °C for several liquids. Comparison with the literature data.

Wavelength (nm)	This work	Coumou et al. (Refs. 28-30)	Leite et al. (Ref. 14)	Finnigan and Jacobs (Ref. 12)	Kaye and Havlik (Ref. 10)	Pike et al. (Ref. 11)	Bender et al. (Ref. 13)
Benzene							
366	9.10						
436	9.93						
488	9.49			10.51			
514.5	9.42						
546		9.78					9.13
633			8.65				
647.1	8.95				9.28	8.81	
Wavelength average	9.29	9.61					
Overall average	9.37 ± 0.54						
Toluene							
488	9.28			8.89			8.77
514.5	9.13						
546		9.21					
633			8.30				
647.1	9.08				9.91	9.84	
Wavelength average	9.16						
Overall average	9.16 ± 0.47						
Carbon tetrachloride							
366		7.44					
436		8.16					
488	8.04			9.24			
514.5	8.08						7.97
546		7.92					
633			8.78				
647.1	8.01				8.75	8.37	
Wavelength average	8.04	7.84					
Overall average	8.25 ± 0.50						
Cyclohexane							
488	12.8						
514.5	12.8						12.5
546		12.1					
633							
647.1	11.9				12.8		
Wavelength average	12.5						
Overall average	12.5 ± 0.4						

determination of the calibration factor $1/\sigma 1 P_0$.

Since the relative uncertainty on scattered power is an order of magnitude lower than that on the calibration factor, the uncertainties on $R_{v,v}$ are estimated from the errors on the calibration factor, amounting typically 1% to 2%.

Since no experimental data are available for $R_{v,v}$ in the literature, and since not all literature values for either R_v or R_u are reported at the same wavelengths and temperature, the comparison is performed as follows.

The expression for the wavelength and temperature dependence of the Rayleigh ratio R_u is[26]

$$R_u = \frac{2\pi^2 kT\beta}{\lambda_0^4}\,(n/\alpha)^2\left(\frac{dn}{dT}\right)^2\frac{6+6\rho_u}{6-7\rho_u}, \tag{7}$$

where k is Boltzmann's constant, T is the absolute temperature, β the isothermal compressibility, n is the refractive index, and dn/dT its temperature derivative and α is the isobaric expansivity.

Both n and dn/dT were measured with a Pulfrich refractometer[27] for the investigated liquids at the different wavelengths. Using the values of ρ_u taken from the literature, and with the aid of Eq. (1) relating $R_{v,v}$, R_v, and R_u the ratio β/α^2 is calculated.

Table IV summarizes the results obtained from experimental data at laser wavelengths and from the values reported by Coumou et al.[28-30] frequently used for calibration purposes.[8]

Glancing at Table IV one sees that there is a scatter of about ± 10% in the literature values. However the reported values agree very well with the overall average values. Especially for toluene and benzene the agreement with the data of Coumou is also excellent.

Finally the values of R_v reported at 488 nm by Finnigan and Jacobs[12] and by Bender et al.[13] are converted into the $R_{v,v}$ form with the aid of Eq. (1). The results are tabulated in Table III.

ACKNOWLEDGMENTS

[1] See, for example, *Dynamic Light Scattering*, edited by R. Pecora (Plenum New York, 1985).

[2] Y. Tsunashima, K. Moro, B. Chu, and T. Y. Liu, Biopolymers 17, 251 (1978).

[3] A. Patkowski, W. Bujalowski, B. Chu, R. Schneerson, and J. B. Robbins, Biopolymers 21, 1503 (1982).

[4] D. Jolly and H. Eisenberg, Biopolymers 15, 61 (1976).

[5] C. Voordouw, Z. Kam, N. Borochov, and H. Eisenberg, Biophys. Chem. 8, 171 (1978).

[6] S. Bantle, M. Schmidt, and W. Burchard, Macromolecules 15, 1604 (1982).

[7] J. P. Kratohvil, G. Dezelic, M. Kerker, and E. Matijevic, J. Polym. Sci. 57, 59 (1962).

[8] H. Utiyama, in *Light Scattering from Polymer Solutions*, edited by M. B. Huglin (Academic, London, 1972), Chap. 4.

[9] W. Kaye and A. J. Havlik, Appl. Opt. 12, 541 (1973).

[10] W. Kaye and J. B. McDaniel, Appl. Opt. 13, 1934 (1974).

[11] E. R. Pike, W. R. M. Pomeroy, and J. M. Vaughan, J. Chem. Phys. 62, 3188 (1975).

[12] J. A. Finnigan and D. J. Jacobs, Chem. Phys. Lett. 6, 141 (1970).

[13] T. M. Bender, R. J. Lewis, and R. Pecora, Macromolecules 19, 244 (1986).

[14] R. C. Leite, R. S. Moore, S. P. Porto, and J. E. Ripper, Phys. Rev. Lett. 14, 7 (1965).

[15] S. H. Maron and R. L. Lou, J. Polym. Sci. 14, 29 (1954).

[16] D. Goring, M. Senez, B. Melanson, and M. Huque, J. Colloid Sci. 12, 412 (1957).

[17] J. Kraut and W. Dandliker, J. Polym. Sci. 18, 563 (1955).

[18] C. I. Carr and B. H. Zimm, J. Chem. Phys. 18, 1616 (1950).

[19] A. K. Van Helden, J. W. Jansen, and A. Vrij, J. Colloid Interface Sci. 81, 354 (1981).

[20] F. A. Johnson, R. Jones, T. P. McLean, and E. R. Pike, Phys. Rev. Lett. 16, 589 (1966).

[21] P. Walstra, Br. J. Appl. Phys. 16, 1187 (1965).

[22] G. F. Lothian and F. P. Chappel, J. Appl. Chem. 1, 475 (1951).

[23] J. K. G. Dhont, Physica A 120, 238 (1983).

[24] R. Finsy, E. Moreels, A. Bottger, and H. Lekkerkerker, J. Chem. Phys. 82, 3812 (1985).

[25] P. Doty and R. Steiner, J. Chem. Phys. 20, 85 (1952).

[26] J. L. Lundberg, E. J. Mooney, and K. R. Gardner, Science 145, 1308 (1964).

[27] E. Moreels, C. De Greef, and R. Finsy, Appl. Opt. 23, 3010 (1984).

[28] D. J. Coumou, J. Colloid Sci. 15, 408 (1960).

[29] D. J. Coumou, E. L. Mackor, and J. Hijmans, Trans. Faraday Soc. 60, 1539, 2244 (1964).

[30] D. J. Coumou and E. L. Mackor, Trans. Faraday Soc. 60, 1726 (1964).