Azimuthal integration of scattered light intensity using a conical lens

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A simple optical scheme using a conical lens to perform azimuthal integration of light collected in a light scattering experiment is described. A substantial increase in the signal-to-noise ratio of the scattered light intensity is achieved. The scheme is particularly useful in nonequilibrium experiments such as spinodal decomposition, where the angular distribution of scattered light varies slowly in time and is peaked in the forward direction.

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INTRODUCTION

In recent years, light scattering measurements have contributed much information concerning the structure of fluids and liquid mixtures near the critical point. Since the characteristic size of the density or composition fluctuations which scatter light typically approaches or exceeds the wavelength of visible light, one is often interested in measuring the angular distribution of scattered light at small angles, i.e., angles less than 20 deg. A variety of techniques have been developed to make angular distribution measurements in binary liquid mixtures near, or slightly below, the critical consolute point.[1,2] In each of these schemes, the light intensity is measured over a range of polar angles θ and at a *single* azimuthal angle ϕ, the polar axis being defined by the incident light beam. A substantial increase in the signal-to-noise ratio could be achieved if, instead of measuring light scattered at a fixed θ and ϕ, the intensity at each angle θ could be integrated over the entire range of ϕ (i.e., 0–2π). We describe here a simple scheme developed in our laboratory to perform such an azimuthal integration which makes use of a conical lens to collect the scattered light.

I. CONICAL LENS OPTICS

The optical arrangement is shown in Fig. 1. A conical lens collects light scattered from the scattering volume, considered here to be a point source. The conical lens is positioned a distance s from the scattering volume so that its axis of symmetry lies along the polar axis. The salient features of the conical lens are: (1) light rays emerging from the scattering volume at different θ are focused at different distances f, from the point of the lens (i.e., the focal length f, is a *monotonic* function of θ), and (2) for a given scattering angle θ, light from the full range of azimuthal angles (0–2π) is collected by the conical lens and focused at a *single point*. Thus, the angular distribution of scattered light integrated over all azimuthal angles can be measured as a function of scattering angles θ, by moving a suitable light detector along the line of focal points (on the previously defined polar axis) of the conical lens. For a given s and θ, a straightforward application of Snell's Law gives

$$f = (\cot \gamma - \tan \beta)\frac{s \tan \theta + t \tan \alpha}{1 + (\tan \alpha)(\tan \beta)}, \qquad (1)$$

where

$$\alpha = \sin^{-1}[(\sin \alpha)/n],$$
$$\gamma = -\beta + \sin^{-1}[n \sin(\beta - \alpha)],$$

n is the index of refraction of the lens glass, β is the cone angle shown in Fig. 1, and t is the thickness of the lens. Equation 1 can be used to design a lens suited to a particular application. The greater the values of n and/or β, the narrower is the range of distances f, over which the scattered

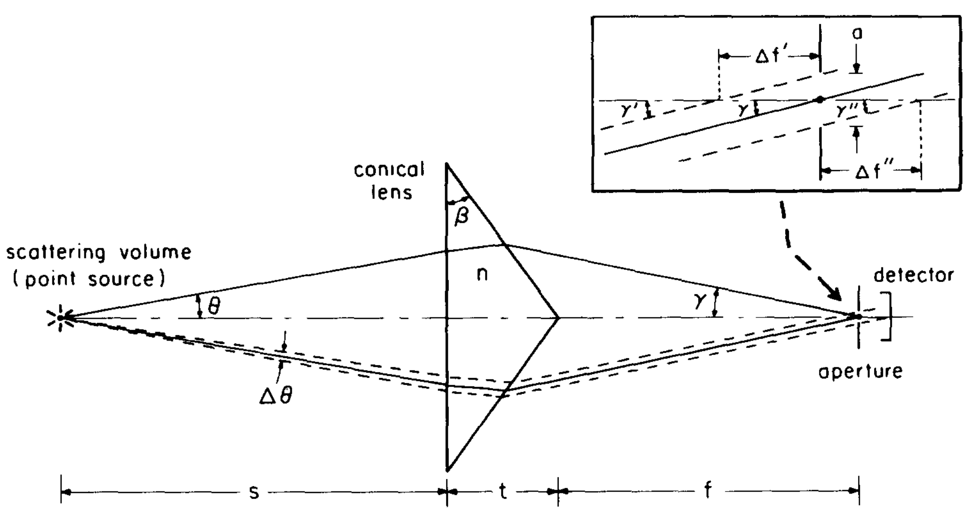

FIG. 1. Optical arrangement for conical lens. The inset in the upper right corner is an enlargement of the aperture and light rays passing through the aperture.

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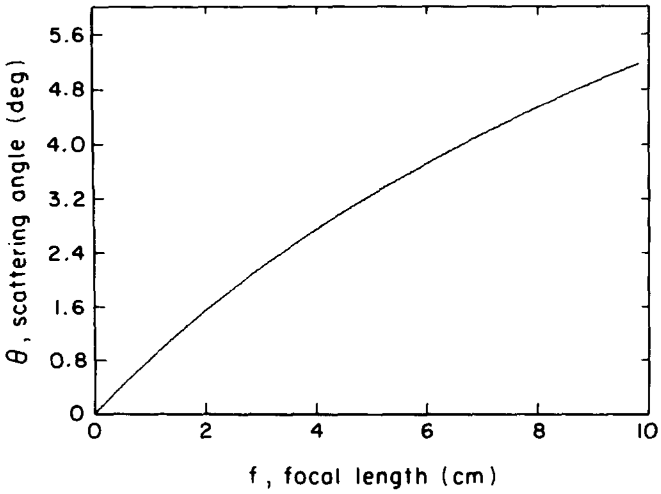

FIG. 2. Plot of scattering angle θ, vs focal length f. The distance from the scattering volume to the lens s, is 22.4 cm.

light is focused. The lens used for this study was made from BK7 glass ($n = 1.518$), with $\beta = 27.5$ deg, and a diameter of 7.00 cm corresponding to a thickness $t = 1.82$ cm. A plot of θ vs f for the above set of parameters is shown in Fig. 2.

An alternative optical arrangement which interposes a spherical lens of focal length s between the scattering volume and the conical lens is shown in Fig. 3. Note that the spherical lens is its focal length away from the scattering volume. The properties of the spherical lens are considered here to be "ideal", that is, well approximated by the thin lens formula. Again, f can be related to θ using Snell's Law

$$f = (\cot \gamma - \tan \beta) s \tan \theta, \qquad (2)$$

where $\gamma = -\beta + \sin^{-1}(n \sin \beta)$. Equation 2 is just the small θ, large s limit of Eq. 1. Using a spherical lens in the manner described above leads to a considerable simplification in the relationship between f and θ. This advantage is offset, however, by the need to make corrections to the thin lens formula for larger angles and by the fact that Eq. 1 reduces to Eq. 2 for small angles, making the introduction of a spherical lens a needless complication.

The angular resolution of the conical lens is determined by the size of circular aperture placed in front of the light detector. The upper right inset in Fig. 1 shows an enlargement of the aperture and light rays. For a given focal length $f(\theta)$, the smallest angle θ', admitted by an aperture of diameter a is given by the condition

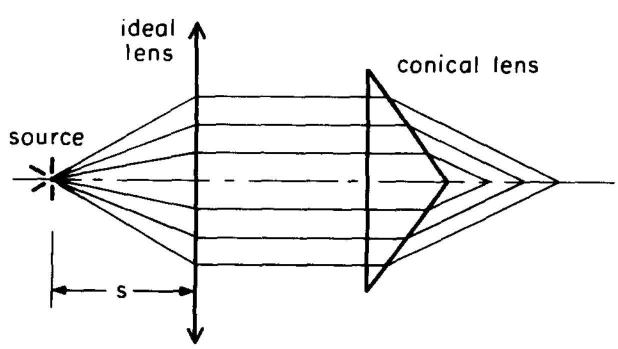

FIG. 3. Optical arrangement for conical lens using a spherical lens. The spherical lens is considered to be ideal; its focusing properties are described by the thin lens formula.

$$\tan \gamma' = a/2\Delta f', \qquad (3)$$

where $\Delta f' \equiv f(\theta) - f(\theta')$. (Note that γ' is a function, albeit weak, of θ'.) Similarly, the largest angle θ'', admitted by the aperture is given by the condition

$$\tan \gamma'' = a/2\Delta f'', \qquad (4)$$

where $\Delta f'' = f(\theta'') - f(\theta)$. Equations 2 and 3 can be solved numerically to obtain the angular resolution $\Delta\theta = \theta'' - \theta'$. Using an aperture of $a = 0.10$ cm with the lens and parameters shown in Fig. 2, the angular resolution varies from 0.28 deg for the smallest θ to 0.17 deg for the largest θ plotted.

The signal measured by the light detector is proportional to the scattered light intensity per unit solid angle multiplied by the solid angle determined from the angular resolution discussed above. The solid angle $\Omega(\theta)$ is given by $\Omega(\theta) = 2\pi(\cos\theta' - \cos\theta'')$, where θ' and θ'' have been determined from Eqs. 3 and 4. Therefore, the light intensity per unit solid angle is obtained by dividing the measured light intensity at each θ by $\Omega(\theta)$.[3]

II. LIGHT DETECTION

The light detector used in this study was an EG & G SD-100 photodiode. Mounted on a translation stage, the photodiode could be positioned anywhere within a range of 0–10 cm along the line of focal points of the conical lens. The translation stage was connected to a stepping motor which allowed the photodiode to be moved periodically and at constant speed along the line of focal points. The output of the photodiode was digitized and sent to a PDP 11/24 computer for storage and processing. In this way, angular distributions of scattered light were quickly collected, one after another.

To protect the photodiode from the unscattered light beam, a small circular piece of tape (diameter = 0.20 cm) was fixed to the center of the flat side of the conical lens. The tape blocked the central beam so that it could not pass through the lens and reach the photodiode.

One concern in using any light detector is its angular response, i.e., how its sensitivity and output vary with the angle the incident light makes with the photosensitive surface. From Fig. 1 it can be seen that the light incident on the detector makes an angle γ with a line normal to the detector surface. In the case where a spherical focusing lens is used (Fig. 3), γ is independent of θ (and, hence, f). In the case where no spherical focusing lens is used, γ is usually a sufficiently weak function of θ so that the variation in detector sensitivity with γ is negligible (see the expression for γ following Eq. 1). For the set of parameters and range of scattering angles in Fig. 2, γ varies between 11 and 17 deg. Of course, significant variation in detector sensitivity with incident angle can easily be taken into account.

III. MEASUREMENTS

Measurements with several light scatterers were made using the conical lens in the arrangement shown in Fig. 1. First, a diffraction grating was used as a scatterer to diffract light from a He–Ne laser (wavelength = λ = 6238 Å). Although the beam of light (diameter 0.15 cm) diffracted by a

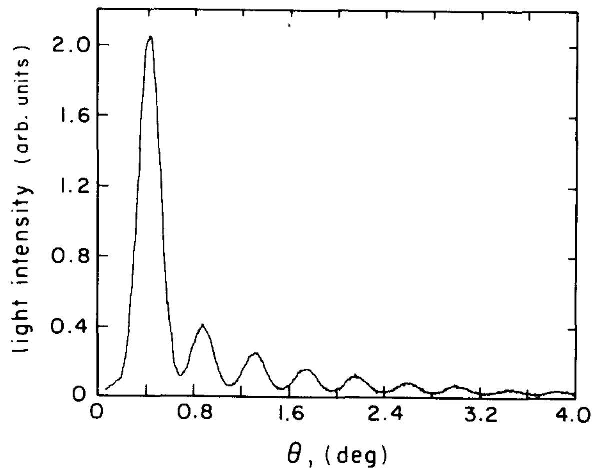

FIG. 4. Light intensity vs scattering angle θ, for 632.8-nm light scattered from a diffraction grating having 118 lines/cm (0.10 cm $< f <$ 9.34 cm, $s = 31.9$ cm). The central peak was blocked by a piece of tape on the flat side of the conical lens (see Sec. II).

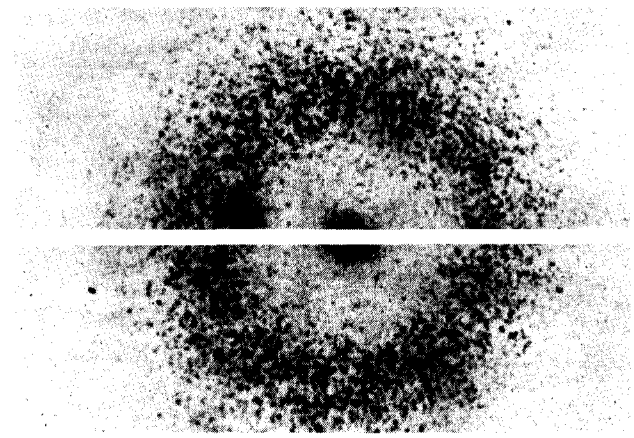

FIG. 5. Photograph of far-field scattering pattern from a critical mixture of methanol and cyclohexane undergoing spinodal decomposition. (Taken from Huang *et al.*[4]) The white line indicates the region over which the light intensity is measured in a typical measurement made with a periodic rotating mirror.[2]

grating does not possess the azimuthal symmetry which makes using a conical lens advantageous, a diffraction grating is useful in aligning the conical lens and verifying Eq. 1.

Figure 4 shows an angular distribution measurement of light scattered by a diffraction grating and collected by the conical lens and detection system described above. Equation 1 was used to convert the distance of the light detector from the conical lens point into a scattering angle. As expected, the angles at which the diffraction peaks occur are consistent with a diffraction grating having $N = 118$ lines/cm (300 lines/in.), i.e., the diffraction peaks are separated by $\Delta\theta \simeq \lambda N = 0.43$ deg.

The diffraction pattern from the grating can also be used to make sure that the lens is properly aligned so that the line normal to the flat side of the conical lens is parallel with the incident light beam. If the lens is slightly tilted away from the proper alignment, the diffraction peaks will broaden. The lens is properly aligned when the widths of the diffraction peaks have been made as narrow as possible.

The lens was also used to collect light from a small incandescent light bulb which served as an approximate point source of isotropic light. For such a source, the light signal recorded by the photodiode should be independent of θ once a correction for the variation in $\Omega(\theta)$ has been made. The results of such a measurement are consistent with this expectation.

The third, and final, scattering source used to test the conical lens system was a binary liquid mixture, of critical composition, undergoing phase separation via spinodal decomposition.[4] Figure 5 shows a photograph of the far-field scattering pattern from such a phase-separating mixture (taken from Huang *et al.*[5]). It should be emphasized here that phase separation via spinodal decomposition is a nonequilibrium time-dependent process. As phase separation proceeds, the characteristic radius of the ring of scattered light (see Fig. 5) typically shrinks on a time scale of minutes. Therefore, each measurement of the angular distribution must be taken over a time interval that is short (i.e., seconds) compared to the characteristic time for the collapse of the ring of scattered light. In a typical experiment using a rotating mirror scheme[2,6] the scattered intensity is measured over a range of polar angles and at a single azimuthal angle as indicated by the white line in Fig. 5. The granular pattern of scattered light, which varies slowly in time, leads to very noisy signals. This is seen in Fig. 6 which shows a measurement made using a rotating mirror.[2] These data were taken from the work of Easwar *et al.*[6] Figure 7 shows a measurement of spinodal decomposition which has been made using the conical lens optical system. Comparing Figs. 6 and 7, one sees that the conical lens leads to a dramatic increase in the signal-to-noise ratio by virtue of the azimuthal integration. The improvement in signal-to-noise is ~ 50.

It should be noted that the noise arising in the measurement of spinodal decomposition is due primarily to temporal fluctuations in the scattered light intensity which are spatially correlated over the entire range of angles measured. Such a situation is peculiar to spinodal decomposition.[7] Generally, such spatial correlations in the temporal fluctuations are not present, and the signal-to-noise ratio can be expected to be even better than that seen in Fig. 7.

In summary, the lens discussed above can produce drastic improvements in the signal-to-noise (for a given data collection time) over that achieved by conventional methods. This optical scheme comes into its own in measurements of

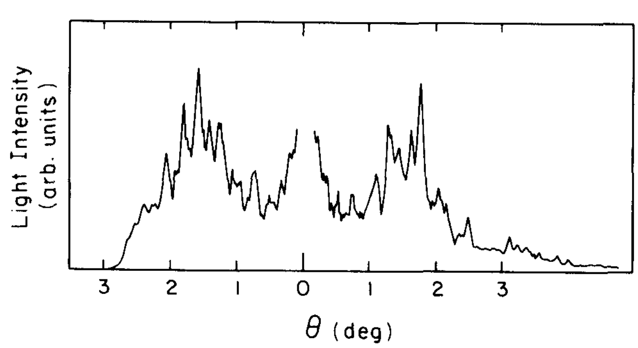

FIG. 6. Angular distribution of light scattered intensity from critical mixture of isobutyric acid and water, undergoing spinodal decomposition. Data obtained using periodic rotating mirror technique (taken from Easwar *et al.*[5]) The angular resolution is approximately 0.2 deg.

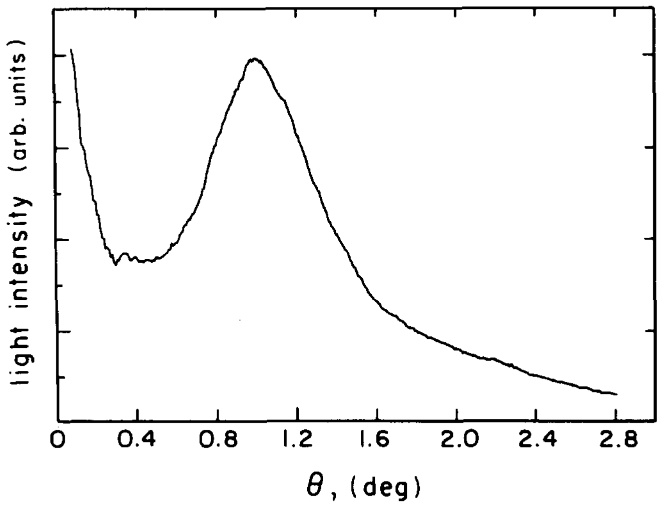

FIG. 7. Angular distribution of light scattered from critcal mixture of isobutyric acid and water undergoing spinodal decomposition. Data obtained using conical lens system described in this study. The angular resolution varies from 0.19 to 0.25 deg over the range of angles for which the light intensity was measured.

the transient type, where data must be acquired quickly, and in measurements where diffraction effects produce a strong (but uninteresting) granularity in the light intensity, as in Fig. 5. Another advantage of using the conical lens is that the corrections needed for the variation in solid angle with scattering angle are well-characterized and can be easily calculated on a computer.[3] Moreover, by mapping a two-dimensional (θ and ϕ) scattering pattern onto a one-dimensional [$f(\theta)$] line of focal points, the light detection scheme is greatly simplified.

Caution is advised in using a conical lens for dynamic light scattering measurements of temporal fluctuations[8] since the detected signal will have been averaged over many coherence areas.[9] Thus, while the average light intensity at the detector increases with a conical lens, the normalized rms amplitude of the fluctuations decreases.

A conical lens is further limited in that it is useful in collecting light scattered only in the forward direction. In practice, this means angles less than approximately 20 deg, depending on the design of the lens.

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[1]N.-C. Wong and C. M. Knobler, J. Chem. Phys. **69**, 729 (1978).
[2]Y. C. Chou and W. I. Goldburg, Phys. Rev. A **20**, 2105 (1979).
[3]Computer programs (in FORTRAN IV) to calculate $f(\theta)$, $\theta(f)$, θ', θ'', and $\Omega(\theta)$ may be obtained from the author.
[4]W. I. Goldburg in *Scattering Techniques Applied to Supramolecular and Non-Equilibrium Systems*, edited by S.-H. Chen, B. Chu, and R. Nossal (Plenum, New York, 1981).
[5]J. S. Huang, W. I. Goldburg, A. W. Bjerkas, Phys. Rev. Lett. **32**, 921 (1974).
[6]N. Easwar, M. Joshua, J. V. Maher, W. I. Goldburg, Physica **118A**, 268 (1983).
[7]M. W. Kim, A. J. Schwartz, W. I. Goldburg, Phys. Rev. Lett. **41**, 657 (1978).
[8]N. C. Ford, G. B. Benedek, Phys. Rev. Lett. **15**, 649 (1965).
[9]E. Jakeman in *Photon Correlation and Light Beating Spectroscopy*, edited by H. Z. Cummins and E. R. Pike (Plenum, New York, 1974).