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Backscattering of Laser Light from Colloidal Silica

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Abstract—Light-scattering experiments gained prominence as potential applications of quantum optics, nonlinear optics, and photon localization. The possibility of the realization of lasing action in random media has created much interest in the study of the coherent structure of the backscattered light from disordered media. Backscattering (BS) studies are carried out to analyze the possibilities of photon localization in colloidal silica. The scattering enhancement is best associated with the density of the scatterers. The width of the BS cone and, hence, the mean-free path is related to the concentration of the medium. The dependence of the photon wavelength on the possible characteristics of the scattering is presented.

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1. INTRODUCTION

Interest in experiments studying the scattering from highly disordered media have increased very much after realizing the possibility of achieving the localization of light. Coherent backscattering (CBS) refers to the enhancement in the intensity of light scattered in the exact backward direction. Moving away from this exact backward direction, phase differences will develop and average out the interference effect. The result is a cone on top of the diffuse background. The width of this cone will be inversely proportional to the mean-free path and, hence, is a measure of the amount of scattering inside the sample. The backscattering cone contains information about the regions that cannot be accessed with normal optical techniques and, hence, is a powerful tool in characterizing any random medium like colloidal suspensions. Since, from the first experimental observation of the coherent backscattering of colloidal suspensions [1], extensive work has been done in this area, most of which are in the strong localization region [2]. Several aspects of light waves make them highly suitable for the study of important localization effects of photons. In the first place, their equations of propagation are well known and the scattering has been extensively studied for almost a century. Furthermore, their vector character adds a new component to the localization problem as the polarization of light assumes importance in observing the phenomenon. The analysis from polystyrene spheres emphasizes the significance of the light polarization and particle-size dependence on CBS [3]. Some recent studies in CBS from liquid crystals have observed systems with larger mean-free paths that we usually encounter in the classical localization studies [4]. The study of the scattering from the Bose–Einstein condensates (BEC) of atoms are also reported to show CBS resulting in very narrow cones of a few milliradians [5, 6]. In the present study, we are concentrating on the backscattering from silica colloids. It is the possibility of achieving photon localization that has evoked our interest in the backscattering of light from random media. A systematic variation in the width of the backscattered cone with the wavelength of the laser beam and the concentration of the medium has been observed.

1.1. Theory

Coherent backscattering is an interference effect observed in multiple scattering [7]. The probability of a scattered wave returning to its point of origin can be well described only by considering the interference effects. In the direction of pure backscattering, the waves travel along the same light path in opposite directions. Scattered lights with time-reversed paths have the same phase and they interfere constructively. This means that the interference will enhance the return probability and, hence, reduces the diffusion coefficient.

Figure 1 shows the scattering geometry of the time-reversed pair of light paths. Moving away from the

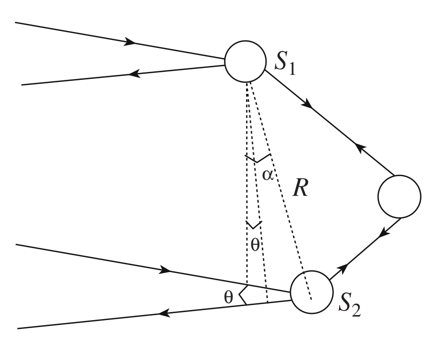

Fig. 1. Scattering geometry.

direction of pure backscattering, the difference in phase $\Delta\phi$ develops, which increases with the angle θ, between the incoming and outgoing wave vectors. Furthermore, this depends on the real-time positions of the first and last scatterers S_1 and S_2 according to

$$\Delta\phi = \frac{2\pi}{\lambda} 2\sin\left(\frac{\theta}{2}\right) R\cos\alpha, \quad (1)$$

where R is the distance between S_1 and S_2. With an increasing θ, the interference effect for an individual light wave will oscillate between the constructive and destructive types. At $\theta = 0$, all time-reversed waves interfere constructively, while at small θ, only light waves with large values of R will interfere destructively and at large θ, the constructive and destructive interferences will average out. Thus, within a certain solid angle around the direction of pure backscattering, there will be an enhanced intensity.

The localization of light in a disordered medium is a transport property affecting the intensity transported through it. The appropriate measure for scattering is the mean-free path length (l) for the light in the medium and the magnitude of the wave vector **k**. The localization is expected for $kl \le 1$, which is known as the Ioffe–Regel criterion [8]. Below $kl = 1$, the electric field cannot even perform one oscillation before the wave is scattered again. Using the photon diffusion approximation [1] for the mean-square separation between the first and last scatterers, Eq. (1) is given as

$$\Delta\phi = \frac{2\pi}{\lambda}\theta\sqrt{2ls}, \quad (2)$$

where $s = ct$ is the total scattering path length of the photon at a random walk, t is the random walk time, and c is the velocity of light propagation [9]. The coherence condition for interference may be stated as

$$\frac{\Delta\phi}{2\pi} \ll 1. \quad (3)$$

The phase coherence is maintained below the critical angle

$$\theta_c \cong \frac{\lambda}{\sqrt{2ls}}, \quad (4)$$

with $s = l$ as the smallest path length,

$$\theta_{\max} \cong \frac{\lambda}{\sqrt{2l}}, \quad (5)$$

which shows that the width of the cone is inversely proportional to the transport mean-free path [10]. One implication for the cone shape is that paths for which the path length is large will only contribute coherently within a relatively small angle about the backscattering direction. That is, the critical angle decreases as the photon path length increases. This implies that physical phenomena such as absorption, sample size, particle density, pump wavelength, and sample reflectivity that affect the scattering paths or the distribution of the path lengths, will manifest itself near the backscattering direction [11–13].

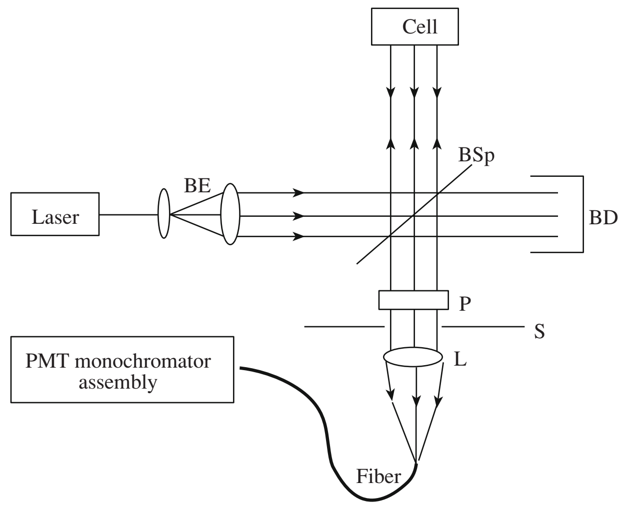

Fig. 2. Schematic representation of the experimental setup. BE—beam expander, BSp—beam splitter, BD—beam dump, S—screen, P—polariser, L—lens, PMT—photomultiplier tube.

The localization effects are present in many random materials. An optimized scattering medium contains scatterers whose size is comparable to the wavelength and highly contrasted refractive index. In order to maximize the randomness in the refractive index, there must be strong density correlations between the scattering centers and a sample with a large volume fraction of scatterers are used. Backscattering experiments that yield a measure of the backscattered cone width and, hence, the mean-free path is essential for understanding and characterizing any disordered medium. Here, we report the backscattering experiments performed on the colloidal suspension of dielectric particles vis a vis silica (SiO_2) with a particle size of 500 nm. The dielectric material has an extremely small optical absorption coefficient in the visible region, and the concentration can be easily varied. In a colloidal suspension, photons will undergo a diffusion-like random walk with an average step size given by the transport mean-free path length (l). Interference between the momentum-reversed paths leads to a cone of enhanced backscattering.

2. EXPERIMENTAL DETAILS

A schematic diagram of our experimental setup is shown in Fig. 2. A linearly polarized laser beam from a diode-pumped solid-state laser (DPSS BWT 50, 40 mW, 532 nm) is allowed to fall on the sample taken in a quartz cuvette. The cuvette is tilted off axis in order to keep its window reflections well away from the detector. The beam divergence is reduced by the expansion of the laser beam [14]. A beam splitter is employed to reflect the light onto the sample. The fraction of the

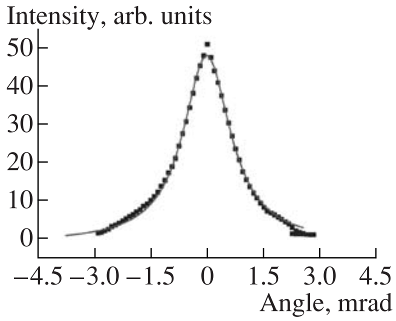

Fig. 3. Backscattered intensity distribution fitted to the Lorentzian profile. Volume concentration of silica colloid is 100%, $P = 40$ mW, $\lambda = 532$ nm.

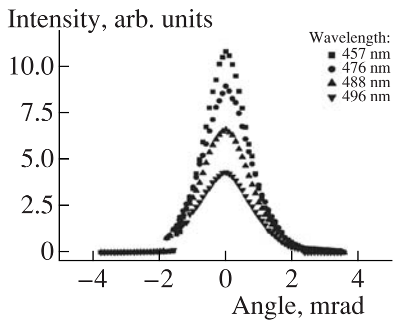

Fig. 4. Variation of the intensity profile with the wavelength: $P = 10$ mW, volume concentration of colloidal silica is 100%.

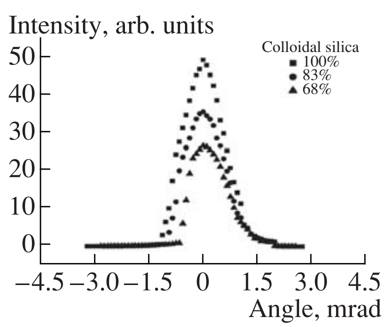

Fig. 5. Variation of the intensity distribution with dilution: $P = 40$ mW, $\lambda = 532$ nm.

original beam that passes through the beam splitter is very carefully dumped since its reflection from the wall would coincide with the 180° backscattered light from the cell. The backscattered light is collected with a proper optical setup. Screens are used to shield all stray radiations. The scattered radiation is measured by collecting it with an optical fiber head, which transports the light to a photomultiplier tube coupled to a 0.2-m McPherson monochromator. An argon-ion laser (Spectra Physics 171 with exciter 270) is used for the study of the variation of the backscattering cone width with the wavelength.

The suspension of SiO_2 particles dispersed in water, obtained from the Aldrich Chemical Company, is used for the investigations. The alteration of the density is affected by the successive dilution with double-distilled water. Samples are homogenized with an ultrasonic bath before being transferred to the cuvette.

A polarizer is placed at the output end of the setup. This is because of the fact that light, being a transverse vector wave, always scatters anisotropically. Thus, the backscattering cones are not necessarily cylindrically symmetric. It consists of components parallel and perpendicular to that of the incident beam and a single sample may yield nonequivalent intensity patterns. One must analyze and detect the incident polarization or its compliment in a backscattering experiment. The choice of the polarization state detected profoundly influences the observed intensity profile. The polarizer provides a polarization-conserving [15] route for the scattered light. This has the added benefit of suppressing the single scattering, which contributes to the incoherent background of the detected signal. The alignment is obtained by replacing the cell with a flat mirror. The positions of the mirror and the beam splitter are alternately adjusted so that the reflected light enters the detector through the lens and the polarizer.

3. RESULTS AND DISCUSSIONS

A systematic study of the dependence of the backscattering intensity profile on the wavelength of the laser beam and the concentration of the colloidal sample is done. The backscattered-intensity distribution shows a typical Lorentzian profile [4] as shown in Fig. 3, which point towards the possible existence of the coherence in the backscattered optical wave. The backscattering of the light from the colloidal suspension ZnO particles of a nanometer size has been reported and it is found to be a good method to characterize and study the colloidal medium [16].

The results in Fig. 4 show a decrease in the intensity of the BS light with an increase in the wavelength of the laser beam and this is in perfect agreement with the usual light scattering in liquids. The scattering of radiation at shorter wavelengths will increase the number of scattering events and, hence, the components of the coherent structure in the BS light. The cone-width distribution also shows a clear dependence on the particle density. The full width at half maximum (FWHM) of the cone decreases with dilution as is evident from Fig. 5. This can be attributed to the effect of an increasing transport mean-free path within the medium. These concentration-dependent studies also provide support to the existence of the coherence in the backscattered intensity profile. The width of the scattering cone is reported to decrease with the concentration of ZnO in DEG as well as the ZnO size in the case of PVP capping

Variation in the cone width of the backscattered beam with the wavelength and percentage volume concentration of colloidal silica

Parameters	FWHM, mrad
Wavelength, nm	
Volume conc. of colloidal silica is 100%*, P = 10 mW	
457	1.41
476	1.63
488	1.72
496	1.88
Volume concentration of colloidal silica, % λ = 532 nm, P = 40 mW	
100	1.41
83	1.37
68	1.21

* 100% volume concentration of colloidal silica means the colloidal silica as received from the Aldrich Company.

[16]. The profile exhibits a Lorentzian one, which characterizes the coherent nature of the BS light. The variation in FWHM of the backscattered cone with the various influencing parameters is given in the table. From the table, it is inferred that an enhancement in the coherent nature of the backscattered light is observed at shorter wavelengths and lower concentrations as indicated by the sharpening of the scattering cone.

4. CONCLUSIONS

The backscattering of light is observed to have a clear dependence on the concentration of the colloidal medium and the wavelength of the laser beam. The possibility of the coherent nature of the backscattered radiation is seen from the Lorentzian profile of the scattering cone. The coherence in the backscattering profile becomes predominant at shorter pump wavelengths and lower concentrations.

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