Progr Colloid Polym Sci (1997) 104:117–120

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Approaching the limits of multiple scattering decorrelation: 3D light-scattering apparatus utilising semiconductor lasers

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Abstract Light scattering as a function of scattering angle can be regarded as a standard method to investigate the dynamics of dilute colloidal suspensions. Concentrated suspensions, which are of interest if interactions between the particles are to be investigated, usually show strong multiple scattering. Decorrelation of multiple scattered light, which isolates single scattering events at the expense of a reduced signal-to-noise ratio, has been proven to work using the two-colour cross-correlation scheme.

In this contribution we demonstrate for the first time the suppression of multiple scattering of a concentrated colloidal suspension at different scattering angles using the 3D cross-correlation technique. Our set-up is designed to extend both measurements towards smaller q values and the scattering intensity of the samples under study beyond the limits of existing apparatus. The latter feature will enable us to approach the photon-diffusion regime as far as possible using decorrelation methods.

Key words Dynamic light scattering – multiple scattering – cross-correlation – colloidal physics

Introduction

Since long there have been theoretical publications to account for the effects of multiple scattering [1–5]. On the other hand, different experimental methods were suggested [6, 7] and proved to work [8–10]. Most recently, the two-colour scheme demonstrated by Drewel et al. is commercially available and its suitability for the investigation of different aspects of colloidal physics has been demonstrated [11]. However, the complexity of this set-up hinders a broad application of multiple scattering decorrelation techniques. We therefore describe in this contribution the 3D set-up, which is by far more easy and stable compared to the two-colour set-up, but offers comparable experimental possibilities.

Experimental details

The experimental set-up is shown in Fig. 1. For a detailed discussion of the "Evolution" of light scattering we refer to [12].

The near IR wavelength ($\lambda = 790$ nm) of the semiconductor lasers guarantees a small scattering cross section and the availability of a comparably low q. The laserdiodes operate under single-mode condition which assures a coherence length of about 1.5 mm, which is large compared to the scattering volume dimensions of about 250 μm. A temperature stabilisation circuit prevents the diodes from mode hopping due to temperature changes. Both lasers share the same wavelength to within $\Delta\lambda = 0.05$ nm, which is necessary for a large intercept in cross-correlation mode [7].

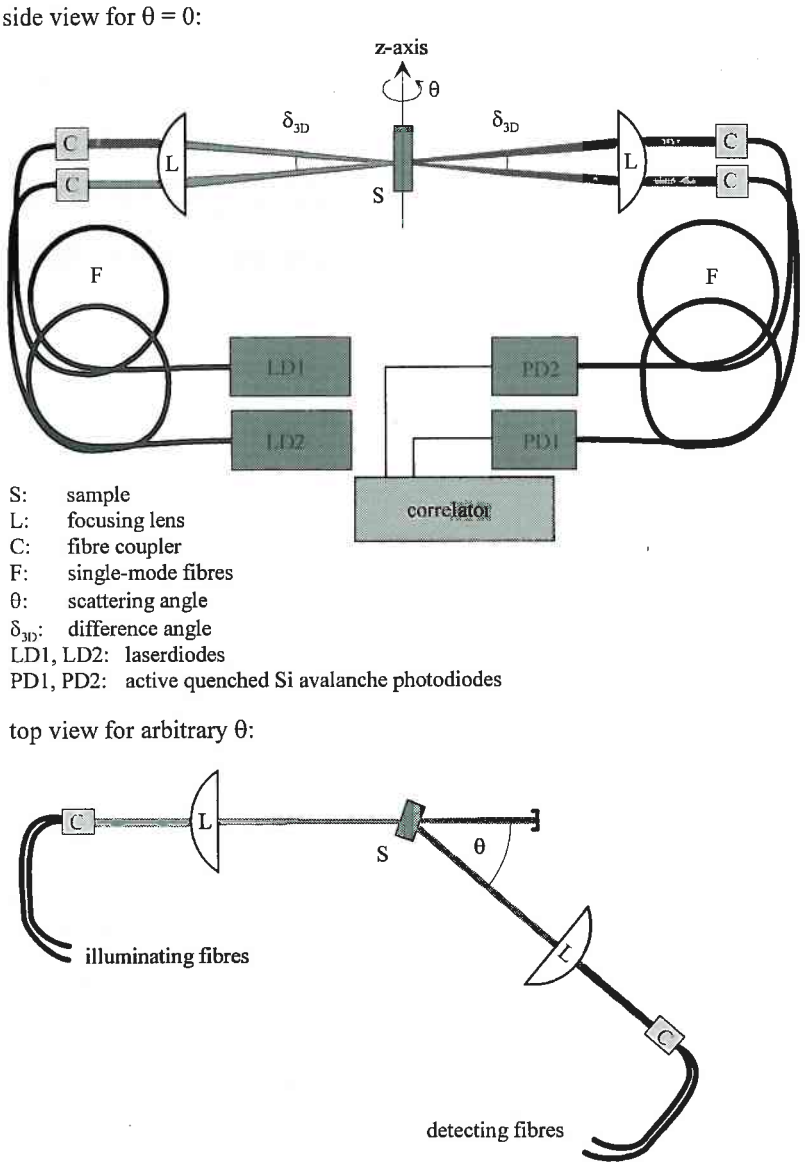

Fig. 1 Experimental set-up of the 3D cross-correlation scheme in side view and top view, respectively

The use of photomultipliers as single photon counters is impossible because of their low responsitivity at the wavelength chosen. Therefore, we use Si avalanche photodiodes as detectors. The active quenching electronics is home-build and quenches the bias voltage periodically ($\nu = 12$ MHz) below breakdown. This leads to an estimated dead-time of the detectors of approximately 40 ns.

Fibre optic components (fibre couplers, single-mode fibres on the detection sides, polarisation maintaining fibres on the transmission sides) as well as the common large lens have been chosen for a maximum compactness and easy alignment of the set-up. On the transmission side, the fibres generate a diffraction limited Gaussian beam out of the transversally multi-mode structure of the laser diode radiation field. On the detection side they enable the maximum intercept possible to be obtained [13].

While the incident beams are polarised (25 dB) perpendicular to the plane of symmetry of the experiment, the scattered light is detected unpolarised. Because the scattering plane in general is inclined with respect to the plane of symmetry, polarisers in the detection optics must be used with care (details will be discussed in a forthcoming publication). In the present investigation by far most of the light is scattered polarised so that the usual V_V geometry may be assumed for the interpretation of the scattering data.

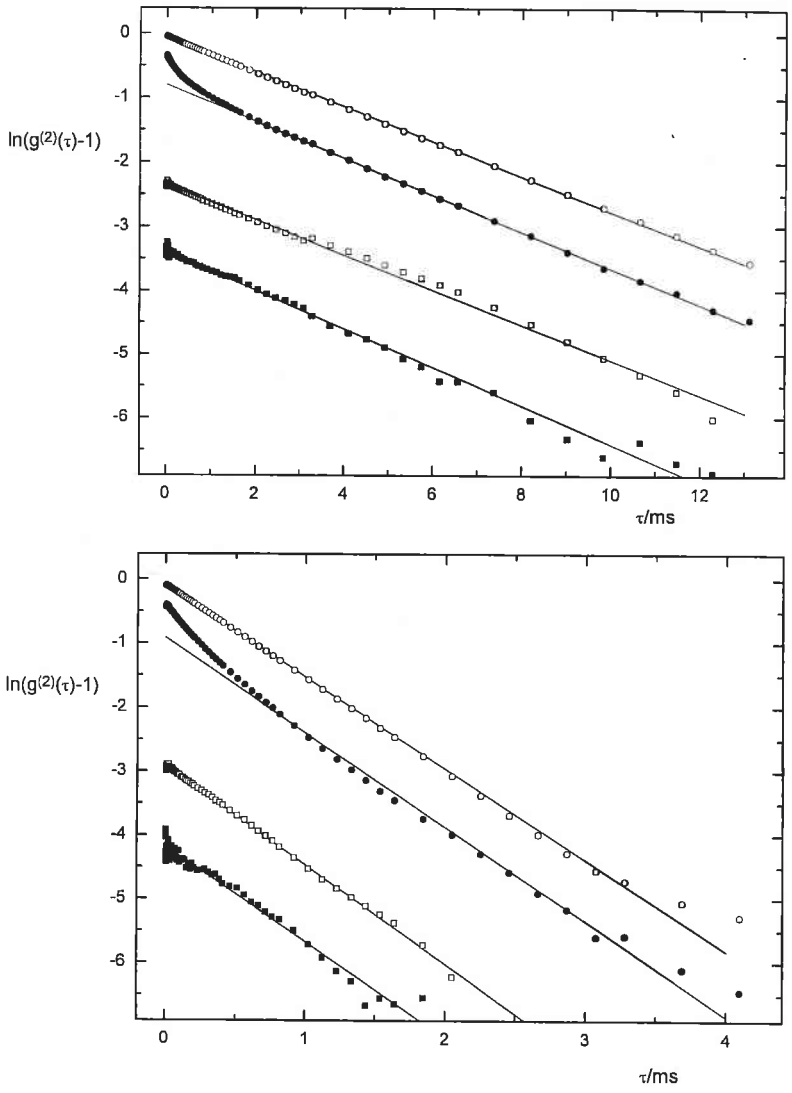

Fig. 2 A) Intensity correlation functions for two polystyrene latex samples ($R = 57.5$ nm) of different turbidity, measured under a scattering angle of $\theta = 36°$ ($q = 6.37\ \mu m^{-1}$); open symbols: $T = 0.69$, closed symbols: $T = 0.02$; circles: auto-correlation mode, squares: cross-correlation mode. B) The same as above, measured under a scattering angle of $\theta = 90°$ ($q = 14.64\ \mu m^{-1}$). For details see text

Results and discussion

Figure 2 show intensity correlation functions determined from polystyrene latex sphere ($R = 57.5$ nm) samples with two different concentrations. The transmission $T = I/I_0$ of the samples was measured to be $T = 0.69$ and $T = 0.02$, respectively. Figure 2A shows the results for a scattering angle of $\theta = 36°$, Fig. 2B those obtained for a scattering angle of $\theta = 90°$ ($q = 6.37\ \mu m^{-1}$ and $q = 14.64\ \mu m^{-1}$, respectively).

In auto-correlation mode, the turbid sample shows a strong distortion at small lag times due to multiple scattering. This distortion can be completely eliminated by cross-correlating the scattered intensities. Fitting either the long-time tail or all data points, respectively, to an exponential decay, the sphere's radius is obtained from all measurements to be $R = 61.6$ nm ($\pm 3\%$), which is in reasonable agreement with the value supplied by the manufacturer (obtained by TEM).

The intercept decreases from auto-correlation mode to cross-correlation mode because of the decorrelation of multiple scattering, which contributes to the background only. However, even under optimal conditions only an intercept of 0.13 has been measured (the theoretical one being approximately 0.40). This unsatisfactory value clearly indicates a necessary improvement of the alignment of the apparatus.

Conclusion

It could be demonstrated for the first time that the proposed innovative cross-correlation scheme, namely the 3D technique, which allows the decorrelation of multiple scattering events in a large angular range, is working as expected. The suitability of multiple scattering decorrelation for the investigation of the physics of strongly scattering colloidal dispersions already has been demonstrated with the two-colour set-up, which is a comparable complicated apparatus. The set-up introduced in this contribution is by far less complex, which might offer the possibility of a broader application of multiple scattering decorrelation techniques.

References

1. Sorensen CM, Mockler RC, O'Sullivan WJ (1976) Phys Rev A 14:1520–1532
2. Sorensen CM, Mockler RC, O'Sullivan WJ (1978) Phys Rev A 17:2030–2035
3. Bøe A, Lohne O (1978) Phys Rev A 17:2023–2029
4. Böheim J, Hess W, Klein R (1979) Z Physik B 32:237–243
5. Dhont JKG, de Kruif CG (1983) J Chem Phys 79:1658–1663
6. Phillies GDJ (1981) J Chem Phys 74:260–262
7. Schätzel K (1991) J Mod Opt 38:1845–1865
8. Phillies GDJ (1981) Phys Rev A 24:1939–1943
9. Mos HJ, Pathamanoharan C, Dhont JKG, de Kruif CG (1986) J Chem Phys 84:45–49
10. Drewel M, Ahrens J, Podschus U (1989) J Opt Soc Amer 7:206–210
11. Segrè PN, van Megen W, Pusey PN, Schätzel K, Peters W (1995) J Mod Opt 42:1929–1952
12. Overbeck E, Sinn Chr, Palberg T, Schätzel K (1997) Colloid Surf A, accepted for publication
13. Ricka J (1993) Appl Opt 32:2860–2875