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# Dynamic light back-scattering with polarization gating and Fourier spatial filter for particle sizing in concentrated suspension

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We present here a new dynamic light scattering method for investigating the particle size in concentrated suspension. By using the polarization gating and Fourier spatial filter, the optical thickness through the sample is reduced and the multiple scattered light is suppressed. The method is systematically tested using suspensions of latex spheres of 100 nm with a wide range of concentrations. Compared to the current common methods for nanoparticle sizing in concentrated suspension, it has the advantage of simple structure and it is easy to debug and maintain.

Keywords: particle sizing, dynamic light scattering (DLS), back-scattering, polarization gating, Fourier spatial filter.

## 1. Introduction

Dynamic light scattering (DLS) has been established as a powerful technique for particle sizing in suspensions over the last decades [1, 2]. The technique takes advantage of the fact that light scattered from a (dilute) suspension of particles fluctuates with a characteristic time scale inversely proportional to the particle diffusion constant. It can provide information about particles with sizes ranging from a few nanometers to several micrometers.

However, DLS applications were only limited to weakly scattering media, in which light propagation can be described by single-scattering models. When it is used for the measurement of undiluted suspensions, the interpretation of a DLS experiment becomes exceedingly difficult for systems with non-negligible contributions from strong multiple scattering. Particularly for larger particles with high scattering contrast this limits the technique to very low concentrations and a large variety of systems are therefore excluded from investigations with dynamic light scattering. Although we can suppress multiple light scattering by sufficiently diluting the system under consideration, in some cases, this may undermine the stability of the original particles and result in failure of the measurement [3].

During recent years, a number of different theoretical and experimental approaches to this problem have appeared. All these methods generally fall into two categories: one is to isolate singly scattered light and suppress undesired contributions from multiple scattering, another is to deduce the particle size information from multiple scattered light where the scattering is so strong that no single scattered light is transmitted. The representatives of the former include: fiber optical quasi elastic light scattering (FOQELS) [4, 5], photon cross correlation spectroscopy (PCCS) [6, 7], low coherence scattering (LCS) [8, 9], and the representative of the latter is diffusing wave spectroscopy (DWS) [10, 11]. These techniques are, however, difficult to implement because of sophisticated mathematical problems or complicated experimental setups.

In this work, we introduce an easy and simple back-scattering setup with polarization gating and Fourier spatial filter techniques which have been well--established and can be easily deployed. In Section 2, we introduce the essential theory of the measurement which includes DLS, polarization gating and Fourier spatial filter technique. In Section 3, a setup of the dynamic light back-scattering (DLBS) and sample preparation are introduced. In Section 4, we compare the measurements of DLBS and DLS and discuss the results. Finally, Section 5 contains concluding remarks.

## 2. Theory

A schematic diagram of the DLBS system is shown in Fig. 1. The intensity correlation function of the scattered light is obtained by measuring the correlation function of the output from a photon multiply tube (PMT) and can be written as:

$$G(\tau) \leq I(t)I(t+\tau) \geq \lim_{T\to\infty} \frac{I}{T}\int_0^T I(t)\, I(t+\tau)\,\mathrm{d}t \tag{1}$$

where $\tau$ is the delay time, $T$ is the total measuring time, $I(t)$ and $I(t+\tau)$ are intensities of the scattered light at $t$ and $t+\tau$, respectively. For monodisperse particles the auto--correlation function (ACF) can be given by:

$$G(\tau) = A\left[1+\beta\exp(-2\Gamma\tau)\right] \tag{2}$$

where $A$ is the baseline of autocorrelation function, $\beta$ is a constant, and $\Gamma$ is the characteristic decay constant which is related to the diffusion constant $D_T$ and scattering vector $q$:

$$\Gamma = D_T q^2 \tag{3}$$

which combined with the Stokes–Einstein relation $D_T = k_B T/3\pi\eta d$, where $\eta$ is the solvent viscosity and $d$ is the diameter, represents the basis for the application of DLS in particle sizing.

According to Rayleigh scattering theory [12], we can see that when linearly polarized light shines a nanoparticle, the backscattered light is also linearly polarized

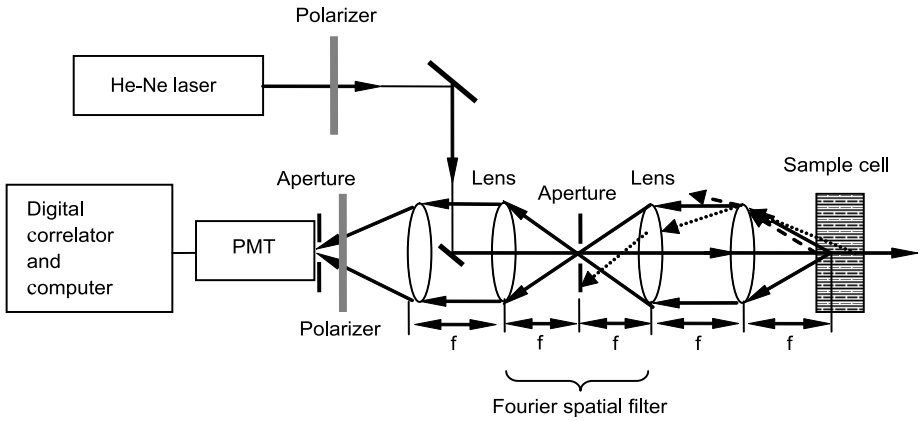


Fig. 1. Schematic diagram of the DLBS setup.

and parallel to incident polarization. But multiple scattered light in a dense random medium randomizes the direction of propagation, phase, and polarization of the incident linearly polarized laser light [13]. Fortunately, the medium typically exhibits significant single scattering, practically in back direction. Any light that is not completely randomized will contain some useful information about the nanoparticle and will be detected, albeit with a low contrast. Therefore, by gating the polarization difference of multiple scattering the single scattered light will be detected.

## 3. Setup

In the setup shown in Fig. 1, a He-Ne laser power is operating at 632.8 nm with a nominal power output of 17 mW. A linear polarizer ensures linear polarization of the input laser. The beam, reflected by a mirror, passes through the Fourier spatial filter (FSF), which is composed of two 100 mm focal-length lenses and a variable--aperture with a diameter of 0.5 mm (the smallest aperture diameter) and acts as a collimator here. And then the light is focused by a lens of focal length $f$ = 100 mm onto a glass sample cell with the focal point designed to fall in a position several microns away from the cell inside wall, which is made with a square base cross-section (10 mm×10 mm).

The backscattered light from the particles is collected by the entire lens. A Fourier spatial filter fixed between the sample cell and the PMT is used to filter out the signals that are not scattered from the focal point. Then the effective scattered light returns to the mirror which blocks the reflected light from the surface of the cell. The polarization state of the backscattered light is discriminated by another linear polarizer whose direction of polarization is parallel to the incident one before the detector. Finally, the single scattered light was gathered into the PMT by the lens, and another variable aperture whose diameter is the same as that of the Fourier spatial filter was set before the PMT to filter the stray light.

To show the ability of our new method to measure the intensity correlation function even in highly turbid samples correct test experiments using suspensions of latex spheres of known size and narrow size distribution were made. The samples we used were aqueous dispersions of spherical latex particles. The sample batch was fairly monodispersed, and its diameter was 100 nm. The original latex suspension has a mass fraction of 10%. The transmission of the particle samples with 100 nm in diameter (mass fraction 10%, 2%, 0.4%, 0.08%) was determined to be 10%, 32%, 66% and 93%.

## 4. Results and discussion

To characterize the system, we compared the $C(\tau)$ (auto-correlation function, ACF) of DLBS and DLS by diluted particle samples of 100 nm. The concentration was chosen to be low enough for multiple scattering contributions to be excluded. Figure 2 shows the delay time $\tau$ dependence of the $C(\tau)$ for this dilute sample. The scattering intensity is sufficiently low, so that the difference between the two curves is rather small but noticeable.

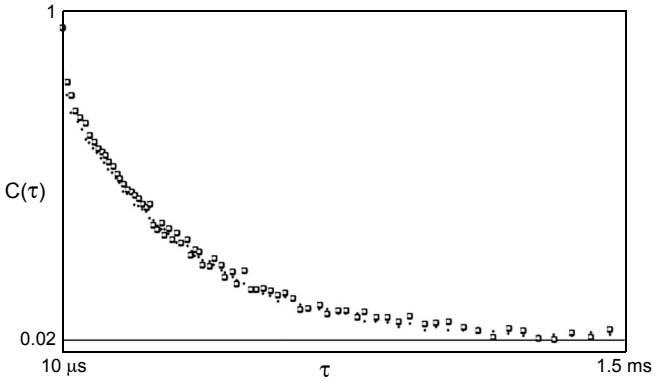


Fig. 2. Comparison of auto-correlation function between DLS (★) and DLBS (□) with the transmissions of 93%, where the abscissa and the vertical axis are both linear coordinates.

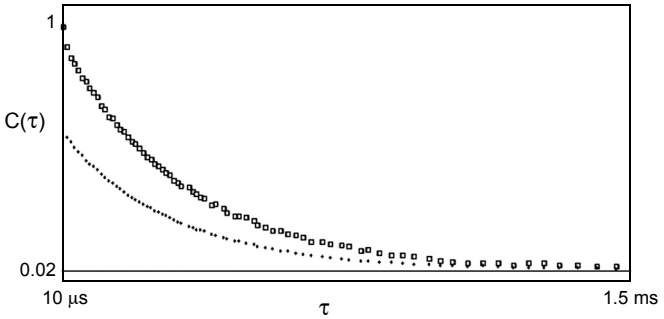


Fig. 3. Comparison of auto-correlation function between DLS (★) and DLBS (□) with the transmissions of 10%, where the abscissa and the vertical axis are both linear coordinates.

The reliability of the DLBS measurement in turbid samples is demonstrated in Fig. 3. The comparison between the ACF of DLBS and of DLS with transmission of 10% is shown. One can see that the ACF of DLS in the highly concentrated sample not only reduces in the amplitude but also decays faster, whereas that of DLBS decays slower.

Furthermore, Fig. 4 shows the variation of the normalized amplitude of the scaled auto-correlation function as the delay time with different transmission. One can see that though the transmission decreases vastly with an increase in concentration, the spread of the decay is acceptable.

Figure 5 shows the curve of measurement results using DLBS and DLS with the decrease of the transmission, respectively. We can see that the result obtained using DLBS is much better than that using DLS. But the error is getting bigger with the decline of the transmission as well.

In addition, the Fourier spatial filter allows only those spatial frequencies that fall below its cut-off frequency to pass through. To study the selection of the variable

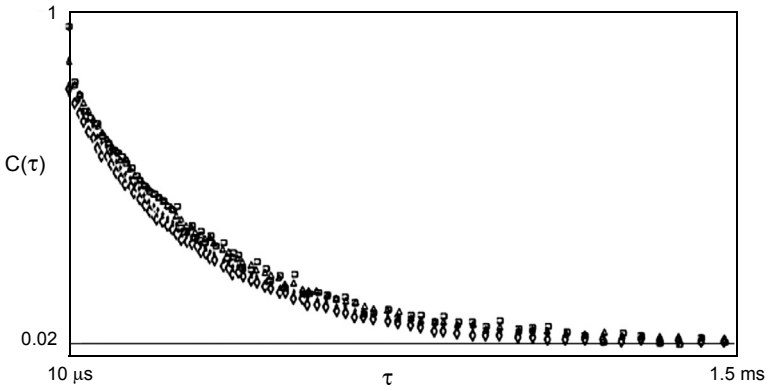


Fig. 4. Variation of the normalized amplitude of the scaled auto-correlation function as the delay time with transmission: □ – 93%, △ – 66%, ★ – 32%, ◇ – 10%, where the abscissa and the vertical axis are both linear coordinates.

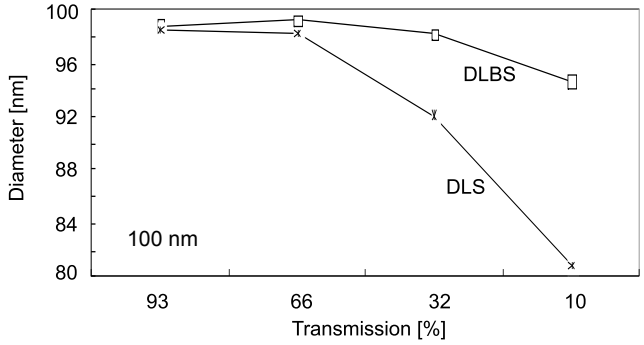


Fig. 5. The diameter measured by using DLBS and DLS as a function of transmission.

aperture size for different concentrations, the measurements were taken with various aperture sizes of 0.5 mm (the smallest aperture diameter), 1 mm and 2 mm at transmissions of 66% and 10%, and variations of the normalized amplitude of the scaled auto-correlation functions as the delay time are shown in Fig. 6. We can see that by reducing the size of the aperture, the ACF of DLBS not only increases in the amplitude but also decays slower which results in the improvement of the measurement. Furthermore, this result is more obvious at high concentration, which is shown in Fig. 6**b**. But the small aperture may also cause the decline of the slick of the ACF. More measurements were carried out with apertures of smaller sizes as 0.1 mm, 0.2 mm, however, the results were not improved. On the contrary,

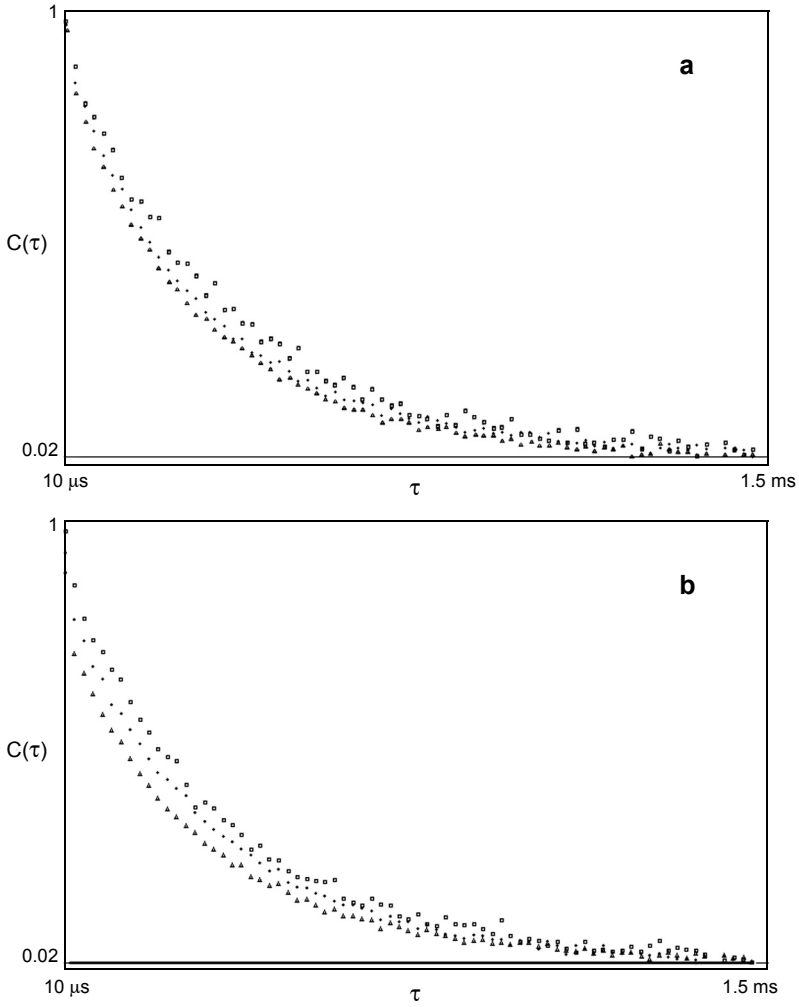


Fig. 6. Variation of the normalized amplitude of the scaled auto-correlation function as the delay time with various aperture sizes of: □ – 0.5 mm, ★ – 1 mm, △ – 2mm at transmission 66% (**a**) and 10% (**b**), where the abscissa and the vertical axis are both linear coordinates.

the adjustment of the optical path is more complex. Therefore, for a certain diameter of particle there should be the best size of aperture for measurement. If the aperture is larger than the best one, the results would deteriorate. On the other hand, if the aperture is smaller than the best one, the adjustment of the optical path would be more complex instead of the enhancement of the results.

## 5. Conclusions

Colloidal suspensions under industrially relevant conditions are frequently turbid and thus very difficult to characterize with traditional dynamic light scattering technique. And the current improved methods are commonly technically extremely demanding and require sophisticated alignment procedures. The dynamic light back-scattering technique not only significantly extends the range of applicability of DLS, but has a simple structure which allows it to be used in more complex working conditions. Systematic experiments have given a good result within a large range of concentration and the potential of this new method as a complementary particle sizing method is very promising.

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