Time-domain depolarization of waves retroreflected from dense colloidal media

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We report on depolarization measurements of femtosecond pulses retroreflected from dense suspensions of silica microspheres with solid loads increasing from 5% to 54%. Backscattered pulse shapes compare well with predictions of the diffusion theory for all volume fractions, and the inferred values of the transport mean free path agree with independent measurements of enhanced backscattering. The measured degree of polarization decays exponentially with temporal rates that scale with the solid load. It is newly found that, for all solid loads, depolarization sets in for path lengths longer than approximately five transport mean free paths.

Light scattering from dense inhomogeneous media has attracted considerable interest over the past decade. Applications beyond medical imaging and biology have benefited from a better understanding of wave propagation and scattering by dense collections of scatterers. In condensed-matter physics, new material-characterization techniques for random media have been developed based on the properties of multiply scattered waves.

Recently, the study of temporal and spatial evolution of the depolarization process has been an area of active research.[1–8] It is widely believed that the depolarization process evolves rapidly when the waves propagate through multiple scattering. However, little is known about the length and time scales of this process and its relation to properties of the random medium. In a previous study,[5] the characteristic depolarization lengths in a medium composed of uncorrelated spherical particles were shown to depend on the size parameter. These experiments were conducted on transmission through slab-shaped media, in which the actual thickness of the medium sets a characteristic length scale. However, applications of industrial interest typically deal with reflection geometries in which the characteristic lengths are determined only by the intrinsic scattering properties of the medium. In this case, no external geometric constraints on the optical wave paths exist. Thus, even simple questions regarding the average photon path length and penetration depth are difficult to answer. Moreover, for media with dense collections of scatterers, the existence of interparticle correlations is expected to play a role in the overall optical properties. For reflection-mode configurations, few experimental measurements have been taken to validate the diffusion-theory-based predictions.

The temporal spread of ultrashort pulses in transmission[9] and in reflection from highly scattering media has been studied recently,[1] and the results were compared with a theory developed for multiple Rayleigh scattering. However, no systematic study of the effects of particle size, volume fractions, or interparticle correlations has been reported so far, to our knowledge. Our goal in this Letter is to estimate the time scales involved in the depolarization process and to study the influence of solid loading of the colloidal system.

We conducted backscattering measurements on colloidal systems with volume fractions ranging from 5% to 54% and consisting of aqueous suspensions of 1-μm-diameter silica spheres with an ionic strength of 0.03 m/L $NaNO_3$ and pH = 9.5. To describe accurately the temporal profile of light pulses backscattered by these dense colloidal systems, we used laser pulses with a duration of 150 fs in a background-free cross-correlation technique.

The laser pulses originate from a dispersion-compensated, self-mode-locked Lexel Ti:sapphire laser pumped by a frequency-doubled Coherent Antares Nd:YAG laser. The Ti:sapphire laser, which had a repetition frequency of 76 MHz, was tuned to a wavelength of 800 nm, and its output was split into two beams by a 50:50 beam splitter, as shown in Fig. 1. One beam passed through a delay stage and served as the gating pulse in the cross correlator. Data runs were typically recorded with a 3-mm (20-fs) step size. The other beam passed through a mechanical chopper, a second beam splitter, and a 15-cm focal-length converging lens to a sample placed at the focus of the beam.

The samples were rotated about the vertical axis so that they were approximately 15° off normal incidence to avoid specular reflections at the inner surface of the quartz container. The degree of polarization of the backscattered light was determined by use of a half-wave plate and a Glan polarizer set to pass vertical polarization only. The polarization extinction ratio for the polarizer was estimated to be better than 1000:1. The backscattered light was cross correlated with the gate (reference) pulse in a $LiIO_3$ second-harmonic-generating crystal. The frequency-

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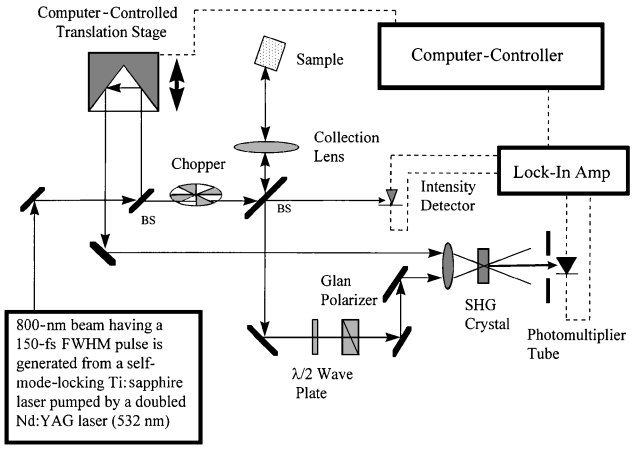

Fig. 1. Experimental setup for measuring the temporal profiles of backscattered light pulses. SHG, second-harmonic-generating; BS's, beam splitters.

doubled light, which was generated only when there was temporal and spatial overlap between the two pulses, was measured by a photomultiplier tube connected to a lock-in amplifier. Typical temporal evolutions of the backscattered intensities are shown in Fig. 2 for both parallel- and perpendicular-polarization channels.

The intensity profiles of backscattered pulses in the two polarization channels are described as a function of number of scattering events by

$$I^{\parallel,\perp}(n) = I(n) f^{\parallel,\perp}(n), \tag{1}$$

where $I(n)$ is the scalar component of the temporal decay predicted by diffusion theory and the factors $f^{\parallel,\perp}$ describe the polarization transfer. For isotropic scattering, these factors are estimated from the Bethe–Salpeter equation as a function of the number of scattering events n (Ref. 10):

$$f^{\perp}(n) = \frac{1}{3}\left[\left(\frac{10}{15}\right)^{n-1} - \left(\frac{7}{15}\right)^{n-1}\right] \tag{2}$$

for the cross-polarized component and

$$f^{\parallel}(n) = \frac{1}{3}\left[\left(\frac{10}{15}\right)^{n-1} + 2\left(\frac{7}{15}\right)^{n-1}\right] \tag{3}$$

for the copolarized component. To evaluate the temporal profile of the backscattered intensity, $I(t) = \sum_{n=1}^{\infty} I(n)c(n,t)$, one needs to know the contribution $c(n,t)$ of the nth-order scattering event to the total light intensity. A Poisson distribution that depends on the value of l^* has been suggested.[1] Using a procedure similar to that used for Ref. 1, we used the pulse profiles in the parallel-polarization channel to infer l^* for samples with different volume fractions, and the results are presented in Fig. 3. For the same colloidal systems, separate measurements of enhanced backscattering were made.[11,12] The angular profile of the coherent enhancement was fitted to a theoretical dependence obtained in the diffusion approximation,[10] and resultant l^* values are also shown in Fig. 3. Both types of measurement reveal the existence of strong interparticle correlations for volume fractions greater than approximately 40%. Values of l^* were also obtained from Mie scattering calculations corrected with the Percus–Yevick structure factor[13] (see Fig. 3). It is worth noting that the Percus–Yevick approximation fails to provide an adequate description of highest volume fractions and that Monte Carlo methods are used at present to improve the understanding of structural correlations in these closely packed media. However, as can be seen from Fig. 3, the l^* values obtained from fitting the temporal profile of backscattered pulse agree well with those inferred from the enhanced backscattering measurements, reflecting the high quality of our data. The resolution of the present temporal measurement allowed us to infer l^* values as small as 20 μm accurately, proving that a structural description of strongly correlated media can be based on temporal analysis of backscattered light pulses.

Alternatively, an absolute measure of the scattering strength or volume fraction of scatterers is provided by the polarization characteristics of backscattered light pulses. We define the degree of polarization as

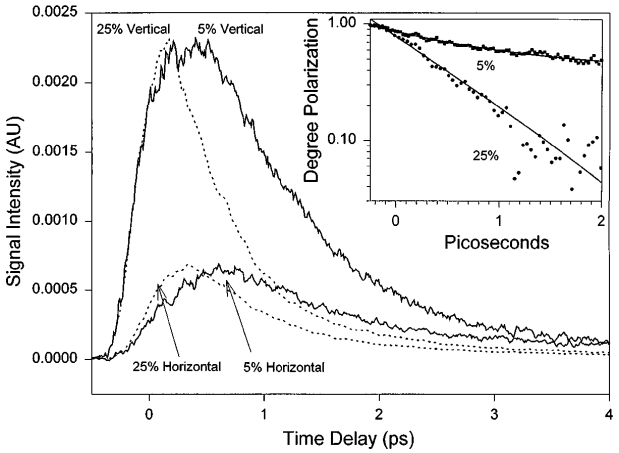

Fig. 2. Backscattered light pulses from dispersions of 1-μm-diameter silica particles with volume fractions of 5% and 25%. The inset shows the temporal evolution of the degree of polarization.

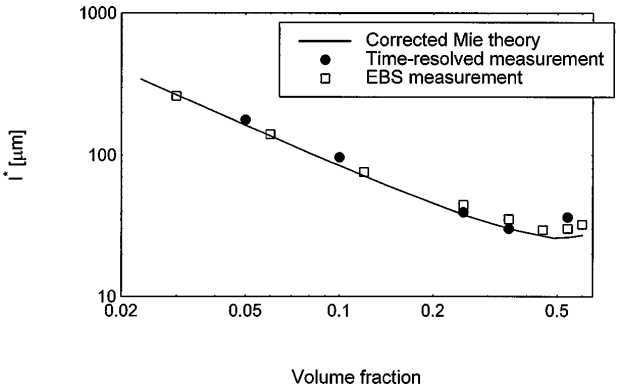

Fig. 3. Values of the transport mean free path corresponding to different volume fractions of 1-μm-diameter silica particles as inferred from enhanced backscattering (EBS) measurements and time-resolved measurements and estimated from Mie theory corrected for interparticle correlations.

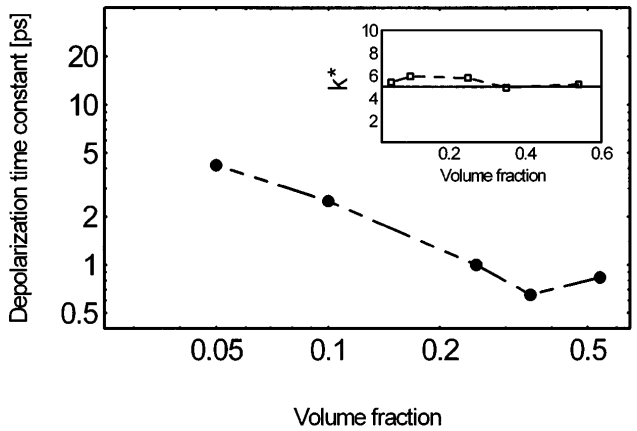

Fig. 4. Depolarization time constants corresponding to dispersions of 1-μm–diameter silica particles with different volume fractions. The inset shows the number of transport mean free paths needed for backscattered photons to become depolarized, as evaluated from the time constant of the depolarization process. Also shown is the theoretical prediction for Rayleigh scatterers.[10]

$$P(n) = \frac{I^{\parallel}(n) - I^{\perp}(n)}{I^{\parallel}(n) + I^{\perp}(n)}, \tag{4}$$

which, if expressed in terms of its temporal evolution, is a directly measurable quantity. Moreover, $P(t)$ does not require knowledge about the probability distributions $c(n,t)$ and results from a relative type of measurement. From Eqs. (1)–(4), it can be shown that the degree of polarization reduces to an exponential dependence on the number of scattering events such as

$$P(n) \approx \exp(-n/\kappa), \tag{5}$$

where κ can be regarded as the average number of scattering events needed to depolarize the optical wave. A simple estimation by use of Eqs. (2) and (3) indicates that κ is $\sim$5. For an effective speed of light, v_{eff}, and an average scattering length l, the time constant of the depolarization process can easily be estimated to be of the order of $\tau = \kappa l / v_{\text{eff}}$.

Using the measured intensities in parallel- and cross-polarized channels, we evaluated the temporal degree of polarization. For the entire set of samples, an initial exponential decay of the degree of depolarization could be followed down to the instrument background level, as can be seen in the inset of Fig. 2. In Fig. 4 the values of the decay constant are presented, and, as can be seen, the decay constant τ gradually decreases when the volume fraction of scatterers increases. However, for the highest density sample, where important interparticle correlation effects are to be expected, τ tends to increase. This can be explained by longer scattering paths and, therefore, by longer traveling times for optical waves in the correlated media. The inset of Fig. 4 shows the values of κ^*, the number of transport mean free paths necessary to depolarize the incident wave. An almost-constant value of 5 is obtained, indicating that, in reflection, photons that have traveled more than $5l^*$ are depolarized. This is a significant result that shows experimentally, for the first time to our knowledge, that temporal evolution of the depolarization process can be treated in a general manner. The estimations of Eqs. (2) and (3) were obtained for Rayleigh scatterers for which the successive scatterings are statistically independent, and, to quote Akkermans *et al.*,[10] they are "by no means universal." The results presented here show that these predictions can be extended to a large size parameter, provided that a renormalization is made from the number of scattering events n to the number of transport mean free paths n^*.

In conclusion, we have systematically measured the co-polarized and cross-polarized temporal profiles of backscattered light pulses and evaluated, for the first time to our knowledge, the temporal evolution of the degree of polarization. We showed that the temporal decay of the degree of polarization can be a direct measure of the solid load of colloidal systems without involving additional assumptions on the path statistics. Moreover, for systems in which l^* ranged from 20 to 200 μm, we found that a constant number of scattering mean free paths is needed for a trajectory to be depolarized in a reflection geometry. As all the characteristics of the multiply scattered light are ultimately dependent on one intrinsic parameter, which is the depolarization factor,[14] structural investigations will benefit from knowledge of the temporal behavior of depolarization phenomena.

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References

1. P. Vreeker, M. P. van Albada, R. Sprik, and A. Lagendijk, Opt. Commun. **70,** 365 (1989).
2. H. Horinaka, K. Hashimoto, K. Wada, and Y. Cho, Opt. Lett. **20,** 1501 (1995).
3. D. Bicout, C. Brosseau, A. S. Martinez, and J. M. Schmitt, Phys. Rev. E **49,** 1767 (1994).
4. A. S. Martinez and R. Maynard, in *Localization and Propagation of Classical Waves in Random and Periodic Structures*, C. M. Soukoulis, ed. (Plenum, New York, 1994).
5. C. Brosseau and D. Bicout, Phys. Rev. E **50,** 4997 (1994).
6. C. Brosseau, Appl. Opt. **34,** 4788 (1995).
7. C. Brosseau, Opt. Lett. **20,** 1221 (1995).
8. M. Dogariu and T. Asakura, Opt. Eng. **35,** 2234 (1996).
9. N. C. Bruce, F. E. Schmidt, J. C. Dainty, N. P. Barry, S. C. W. Hyde, and P. M. W. French, Appl. Opt. **34,** 5823 (1995).
10. E. Akkermans, P. E. Wolf, R. Maynard, and G. Maret, J. Phys. (Paris) **49,** 77 (1988).
11. A. Dogariu, G. Boreman, and M. Dogariu, Opt. Lett. **20,** 1665 (1995).
12. A. Dogariu and G. Boreman, Opt. Lett. **21,** 1718 (1996).
13. J. M. Ziman, *Models of Disorder* (Cambridge U. Press, London, 1979).
14. I. Freund and E. Berkai, J. Opt. Soc. Am. A **8,** 1559 (1991).