# FEMTOSECOND TIME-RESOLVED MEASUREMENTS OF WEAK LOCALIZATION OF LIGHT

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Time-resolved backscattering of coherent light from a disordered medium under weak localization conditions has been observed for the first time with femtosecond resolution using a light-gating technique based on second-harmonic generation. The time dependence of the observed enhanced backscattering is consistent with simple diffusion theory.

One of the interesting aspects of light propagation in disordered media is the possibility of photon localization in analogy with localization of electrons in random metallic systems [1–3]. A first indication of such behavior is the recent observation of weak localization of light [4–7]. Weak localization arises from constructive interference between scattered light waves traveling the same path but in opposite direction. The effect manifests itself as a narrow peak in the intensity of light scattered from a disordered medium in the backward direction (i.e. the direction opposite to the incident one). The lineshape of the coherent backscattering peak and its dependence on the various time-reversed paths was studied previously from CW light scattering experiments. Van Albada et al. [4,8,9] and Etemad et al. [10] studied the variation of the lineshape with the path length of the scattering events. The path length was adjusted by varying the thickness of the sample; for any path that extends beyond the thickness of the sample the photon is eliminated and does not contribute to the backscattering peak. Etemad et al. [10] performed absorption experiments, in which the variation of the lineshape function with the inelastic scattering length in the medium was studied. Photons that take light paths longer than the inelastic scattering length are absorbed and do not give rise to enhanced backscattering.

In CW light scattering experiments it is only possible to study scattering events with a large distribution in path length. In this Letter we report results from coherent backscattering experiments performed in the time domain, with femtosecond time resolution. These measurements allow us to study the contribution to the backscattering interference peak of light trajectories with a specific path length. Very recently, time-resolved light scattering experiments on a picosecond time scale were performed by Watson et al. [11], but no effects of interference could be detected.

The sample used in the backscattering experiments was a 6.8 vol% suspension of $TiO_2$ particles (mean diameter 0.22 μm) in 2-methylpentane-2,4-diol. The elastic mean-free path, $\lambda_{mf}$, in the sample was determined from transmission measurements to be 2 μm [9]. Time-resolved backscattering measurements were performed using a light-gating technique based on second-harmonic generation (SHG) [12]. This technique allowed detection of backscattered pulses with a temporal resolution of 30 fs. In fig. 1 the experimental setup is shown. A colliding-pulse mode-locked (CPM) ring laser operating at 620 nm delivered laser pulses with a duration of 100 fs, a repetition rate of 95 MHz and an average power of 20 mW. The femtosecond laser pulses are split into two pulse trains, of which one is sent into a variable delay line, while the other one is focused by an achromatic lens to a 50 μm spot on the sample sur-

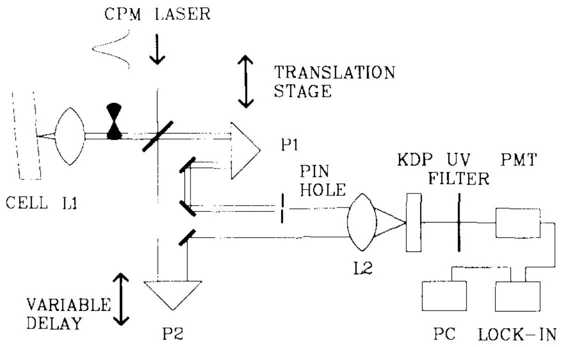


Fig. 1. Experimental setup used in the time-resolved backscattering measurements.

face. Light scattered from the sample is collected by the same lens and focused non-colinearly with the delayed femtosecond pulse onto a KDP crystal. The 5 mm thick crystal is angle tuned for second-harmonic generation. The second-harmonic radiation was passed through an UV-transparent filter and detected by a photomultiplier. To allow lock-in detection of the photomultiplier output, the laser light reflected from the sample was mechanically chopped.

The angular dependence of the scattered light intensity at fixed delay times after the laser flash was measured as follows. The light was sent via a retroreflecting prism, mounted on a translation stage to allow for displacement parallel to the sample surface, onto a pinhole. By translation of the prism, the scattered light beam is displaced parallel to itself, without introducing additional time delay. As the scattered light beam is displaced, different parts of the beam pass the pinhole. In this way, the intensity profile of the beam, which corresponds to the angular distribution of the backscattered light, can be recorded. The fixed position of the pinhole guarantees a constant second-harmonic generation efficiency.

Experimental backscattering peaks obtained at various delay times after the laser flash are presented in fig. 2. The backscattering peaks were detected for light polarized parallel to the polarization of the incoming laser pulse. We found that the width of the peak is reduced from approximately 80 mrad at 30 fs delay to approximately 25 mrad at a delay time of 330 fs. For short delay times large intensity fluctuations were observed in the backscattering peaks which could be reproduced in different scans. The fluctuations arise from additional interference (speckle) of the light scattered from $TiO_2$ particles near the surface of the sample. Apparently, these particles are not subject to rapid brownian motion and no averaging of the speckle pattern occurs. The fluctuations in the scattered light intensity were reduced by spinning the sample. In fig. 3 time-resolved intensity profiles of light pulses scattered from the $TiO_2$ sample are shown. The profiles were detected for the parallel component of the light pulses and obtained at fixed angles with respect to the backscattering direction.

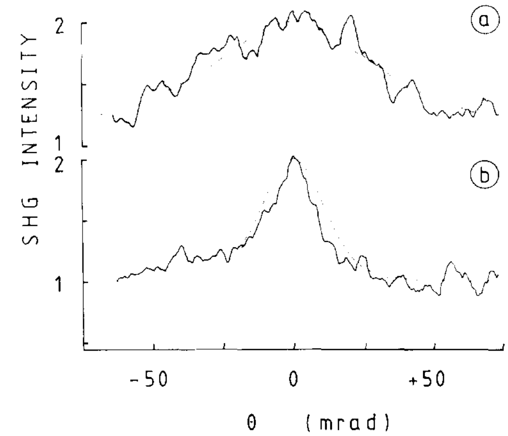


Fig. 2. Enhanced backscattering cones measured for a 6.8 vol% suspension of $TiO_2$ particles in 2-methylpentane-2,4-diol ($\lambda_{mf}=2$ μm) at different delay times after the laser pulse: (a) 30 fs; (b) 330 fs. Dashed curves represent results of calculations based on eq. (2).

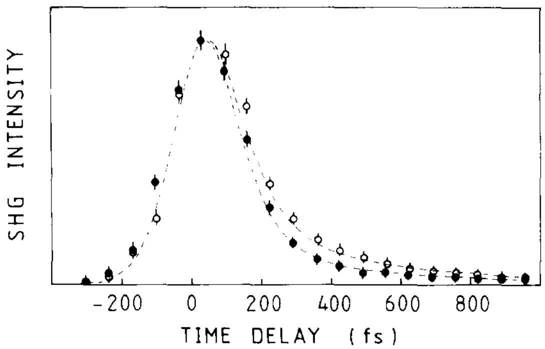


Fig. 3. Backscattered light pulses from a 6.8 vol% suspension of $TiO_2$ particles in 2-methylpentane-2,4-diol ($\lambda_{mf}=$ 2 μm) measured for $\theta=0$ mrad (open circles) and $\theta=27$ mrad (full circles). Dashed curves represent results of calculations based on eq. (2).

The recorded intensity profiles are normalized to unity at zero delay time. The observed backscattered pulses are characterized by a short rise time, which is determined by laser-pulse duration, and a slower long-time decay. The decay could be monitored for delay times up to 10 ps. Photons detected at these long delay times have traveled more than 2 mm in the sample and are elastically scattered over more than one thousand times.

To explain the results qualitatively we consider two time-reversed paths with $n$ elastic scattering events. The total phase shift between the lightwaves which are scattered by the same centers but in reversed sequence equals $(\boldsymbol{k}_i+\boldsymbol{k}_f)\cdot(\boldsymbol{r}_1-\boldsymbol{r}_n)$, where $\boldsymbol{k}_i$ and $\boldsymbol{k}_f$ represent the incoming and outgoing wavevectors and $\boldsymbol{r}_1$ and $\boldsymbol{r}_n$ are the positions of the first and last scattering centers. In the case of backscattering ($\boldsymbol{k}_f=-\boldsymbol{k}_i$) both waves have the same amplitude and phase and add coherently. If $\theta$ is the angle between $\boldsymbol{k}_i$ and $\boldsymbol{k}_f$, the coherence is lost for angles $\theta$ larger than $\lambda/(|\boldsymbol{r}_1-\boldsymbol{r}_n|)$, where $\lambda$ is the wavelength. If the path length associated with the $n$th order scattering event is $L_n$, $|\boldsymbol{r}_1-\boldsymbol{r}_n|$ is approximately $(L_n\lambda_{\mathrm{mf}})^{1/2}$ as follows from simple random-walk arguments, and enhanced backscattering will occur within a narrow cone of angular width $\lambda/(L_n\lambda_{\mathrm{mf}})^{1/2}$. Due to the $L_n^{-1/2}$ factor, scattering events with long light paths will have narrower cones associated with them, in agreement with the experimental results presented in fig. 2.

It is seen from fig. 3 that for all delay times the scattered light intensity is largest at exact backscattering conditions, as could be expected. Under exact backscattering conditions, all time-reversed paths will interfere constructively. The intensity equals almost twice the average scattering intensity. At small angles with the backscattering direction, the phase difference between time-reversed paths increases with $t^{1/2}$ and a coherent contribution to the scattered intensity will only occur during a short time after the laser flash.

We will now give a quantitative analysis of the data presented in figs. 2 and 3. Using the diffusion theory of Akkermans et al. [13,14], we can write for the time dependence of the lineshape function, $\gamma(\theta, t)$, of the coherent backscattering peak (or bistatic coefficient):

$$\gamma(\theta,t)\propto t^{-1/2}[1-\exp(-8.7\lambda_{\mathrm{mf}}/ct)] \times\{1+\exp[-(4/3\pi^2)\lambda_{\mathrm{mf}}ct\lambda^{-2}\theta^2]\}\,, \qquad (1)$$

where $c$ is the speed of light in the medium ($c=c_0/n$, with $c_0$ the speed of light in vacuum and $n=1.51$ is the refractive index in the medium). Eq. (1) describes a gaussian-shaped backscattering peak with width of order $\lambda/(\lambda_{\mathrm{mf}}ct)^{1/2}$. In principle, eq. (1) is valid for times $t$ long enough for the propagation to have reached the diffusion limit, which is only after many scattering events. However, as we will see, it also gives an accurate description for our experiments at short delay times.

To infer the observed lineshape function, $\hat{\gamma}(\theta, t)$, the results should be convoluted with the laser pulse shape:

$$\hat{\gamma}(\theta,t)=\int_{-\infty}^{\infty}\mathrm{d}\tau\int_{-\infty}^{\tau}\mathrm{d}t'\,\gamma(\theta,\tau-t')I_{\mathrm{p}}(t')I_{\mathrm{p}}(\tau-t)\,. \qquad (2)$$

In eq. (2) the first integral gives the backscattered signal as a convolution of the impulse response function of the scatterer, $\gamma(\theta, t)$, and the incident pulse $I_{\mathrm{p}}(t)$. The second integral in eq. (2) gives the measured signal intensity as a convolution of the backscattered signal with the light-gating pulse. Coherent backscattering lineshapes were calculated using eq. (2) and compared with the experimental results in fig. 2. In the calculations a secant hyperbolic pulse shape was assumed with 100 fs duration (fwhm). As is seen from fig. 2, the calculated lineshapes are in good agreement with the experimental data. Calculated pulse shapes based on eq. (2) using fixed $\theta$ values are shown as dashed curves in fig. 3. As is seen from fig. 3 the calculated and experimental pulse shapes are also in good agreement, even for short delay times.

We will now briefly comment on the polarization of the pulses reflected from the sample. For the parallel and perpendicular polarized components of the reflected laser pulse, time-resolved intensity profiles have been recorded. The intensity profiles were obtained outside the narrow cone of enhanced backscattering so no coherent contribution to the scattered intensity was monitored. The results show that in the backscattered light the component with polarization parallel to the polarization of the incident pulse is dominant up to delay times of 500 fs. At longer delay times the scattered light contains equal amounts of the polarized and depolarized components. The time needed to attain complete scrambling of the polarization corresponds to more than 50 scattering events. The experimental result that many scattering events are needed to attain complete scrambling of polarization is in agreement with earlier predictions based on calculations on multiple Rayleigh scattering [15].

In summary, our experiments are the first observations in the time domain of the coherent backscattering effect for waves propagating in disordered media. Both the width of the backscattering peak and the temporal intensity profiles of backscattered pulses are in good quantitative agreement with the theoretical predictions based on simple diffusion theory.

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