Optical path-length spectroscopy of wave propagation in random media

G. Popescu and A. Dogariu

Center for Research and Education in Optics and Lasers, University of Central Florida, Orlando, Florida 32816

Received November 20, 1998

We propose optical path-length spectroscopy as a new approach to obtaining information from media that exhibit multiple light scattering. By using a backscattering technique based on low-coherence interferometry, we are able to determine the optical path-length distribution for light reflected from a random medium and to infer the value of the transport mean free path. We illustrate how a diffusion approximation model leads to a satisfactory description of depth-resolved profiles of the backscattered intensity and discuss potential applications of this technique.

OCIS codes: 260.0260, 290.0290.

Low-coherence inteferometry (LCI), which was developed initially in the field of fiber optics,[1] has become a widely used technique for various applications that involve biomedical imaging.[2,3] The use of light sources with short temporal coherence produces the depth resolution needed for optical imaging, and the method is generally referred to as optical coherence tomography. So far, LCI has been used as a filter that suppresses multiple light scattering and preserves the single-scattering component characterized by well-defined scattering angles and polarization.[3] In this Letter we show, for the first time to our knowledge, that LCI can also be used to investigate the multiple-scattering regime of wave propagation, and we discuss some characteristics that make this approach particularly appealing for applications.

A multiple-scattering regime is usually associated with wave propagation through optically dense random systems and is commonly described in terms of a diffusion equation. This is an approximation for energy transport in which isotropic elastic scattering and wave propagation at a constant group velocity are considered and polarization and interference effects are neglected.[4] Diffusive wave propagation depends on the specific scattering geometry and is characterized by the probability density $P(s)$ of optical path lengths through the medium. In general, $P(s)$ can be theoretically estimated for different experimental configurations, but, so far, direct experimental studies of optical path-length distribution have been limited to investigations of temporal broadening of short light pulses that propagate diffusively.[5–9] The time t necessary for the optical wave to propagate along a path of length s is simply given by $t = s/v$, where v is the average velocity of energy transport. We can relate the steady-state transport mean free path (l_t) to the dynamic diffusion coefficient D by considering a constant energy transport velocity $v = 3D/l_t$. In steady-state conditions, l_t depends on both the number density of the scatterers and the size and shape of each individual scatterer: $l_t = [n\sigma_s(1 - g)]^{-1}$, where n is the number density of scatterers, σ_s is the cross section of a single scattering event, and g is the average cosine of the scattering angle. Note that this definition is valid for random media that are far from the localization condition, i.e., $l_t \gg \lambda$. For media of finite thickness, the condition under which the diffusion theory is generally valid is $l_t/L \ll 1$, where L is the thickness of the random medium. The accuracy of the diffusion approximation has been questioned in the past, and experiments on time-resolved diffuse transmission have shown that it becomes increasingly less reliable when the thickness of the sample decreases and the anisotropy factor increases.[8] However, it has been shown that, when internal reflections at the boundary and scattering anisotropy are properly taken into account, the diffusion predictions are accurate for samples as thin as $\sim 5l_t$.[10] Recently Lemieux *et al.* expanded the limits of applicability of the diffusion approach by using a telegrapher equation that takes into account the ballistic transport and by modeling the scattering anisotropy in terms of a field concentration discontinuity at the source point.[11]

Here we propose a novel approach to investigation of wave propagation through random media. Based on the LCI principle, the new technique, called optical path-length spectroscopy (OPS), directly infers the path-length distribution $P(s)$ of waves scattered by a random medium. In this Letter we present OPS experiments in a backscattering geometry that is appealing for a variety of applications, but this is by no means a restriction on the principle of the proposed method. It is worth noting that the information provided by OPS is somewhat similar to that obtained from time-resolved reflectance measurements, for which the diffusion approximation provides a reasonable description of the experimental data.

In the present LCI geometry, light from a broadbandwidth source is first split into probe and reference beams, which are both retroreflected from a targeted scattering medium and from a reference mirror, respectively, and then subsequently recombined to generate an interference signal. Assuming quasi-monochromatic optical fields ($\Delta\lambda/\lambda \ll 1$), the detected intensity has the simple form $I_d = I_s + I_{\text{ref}} + 2\sqrt{I_s}\sqrt{I_{\text{ref}}}\cos(2\pi\Delta s/\lambda)$, where I_d, I_s, and I_{ref} are the detected, scattered, and reference intensities, respectively. The optical path difference between the scattered and the reference fields is denoted Δs, and λ is the central wavelength. Two conditions must be

0146-9592/99/070442-03$15.00/0

fulfilled to yield interference maxima: Δs must be a multiple of the wavelength, and $|\Delta s| < l_{\mathrm{coh}}$, where l_{coh} is the coherence length of the source. In the present OPS configuration, I_s corresponds to the reflectance of a multiple-scattering medium. In LCI, only the class of waves that have traveled an optical distance that corresponds to the length of the reference arm is able to produce fringes and is, therefore, detected. In the optical path-length domain, the interferometer acts as a bandpass filter with a bandwidth given by the coherence length of the source. Accordingly, the shorter the coherence length, the narrower the optical path-length interval of backscattered light that will produce detectable fringes. Now, if we let the reference mirror sweep the reference arm, waves with different optical path lengths through the medium are detected, and an optical path-length distribution is reconstructed. As the reference mirror is moved, the peak of the beat signal produced is proportional to the squared root of the optical reflectance, the quantity of interest in our backscattering geometry. The interference signal is the dot product of the optical fields returning from the reference and the test arms, and, although the source sends an unpolarized signal into the random medium, polarization effects still occur as a result of the scattering processes. To achieve a polarization-independent measurement we split the unpolarized light coming from the medium into two orthogonally polarized components, which are directed to two photodiode detectors; the reference field is maintained linearly polarized and is equally divided between the two detectors.[12] The amplitude of the beat signal envelope from each detector is measured, the squares of these are added, and the interference signal obtained is independent of the polarization state returning from the scattering medium. A dynamic range better than 90 dB is obtained in practice. We note that the OPS approach is experimentally limited by the fact that the signal that corresponds to long paths within the medium is weak, and therefore a large dynamic range is needed for accurate measurements in the tails of the path-length distributions. However, unlike for dynamic techniques, there is no need for sophisticated time-of-flight configurations, and the measurement can be taken over longer periods of time to yield a better average. The reflectance signal is typically averaged over 100 successive scans, which significantly improves the signal-to-noise ratio.

In this Letter we present OPS data on suspensions of polystyrene microspheres in water, with various volume concentrations. The low-coherence interferometer used for our studies has a light-emitting diode as a source with a coherence length of 10 μm and a broad wavelength band centered at 1300 nm. The same single-mode optical fiber is used to send and collect the light from the random medium. Throughout our experiments, the dimensions of the samples are much larger than the transport mean free path, and the illumination geometry is such that the scattering system can be treated as a semi-infinite medium.

In this particular configuration of OPS, the source and the detector physically overlap, and the incident beam is narrow, collimated, and normal to the wall of the glass cuvette that holds the homogeneous medium. We calculate the diffusively backscattered energy flux by applying a time-dependent diffusion approach. In media with negligible absorption, the diffuse energy density $\Psi(\mathbf{r}, t)$ satisfies the diffusion equation $\partial\Psi(\mathbf{r}, t)/\partial t = D\nabla^2\Psi(\mathbf{r}, t) + \delta(\mathbf{r})\delta(t)$, where $\delta(\mathbf{r})\delta(t)$ is the impulse source at times $t = 0$ and $r = 0$ and D is the diffusion coefficient, given by $D = (vl_t)/3$. Setting an appropriate boundary condition such that Ψ vanishes on the plane $z = -z_0$, we can solve the diffusion equation by using the image source method (see, for instance, Ref. 6), and the energy flux is obtained from Fick's law.[13] Assuming an average energy transport velocity v, the path-length dependence of the energy flux detected in the particular OPS geometry ($r = 0$) can be evaluated to be

$$J(s) = (4\pi l_t/3)^{-3/2} z_0 v s^{-5/2} \exp\left(-\frac{3{z_0}^2}{4sl_t}\right), \qquad (1)$$

where the extrapolation length $z_0 = 2/3(1 + R_{\mathrm{eff}})/(1 - R_{\mathrm{eff}})l_t$ carries the information about the effective reflectivity R_{eff} at the boundaries. The extrapolation length ratio z_0/l_t is critical for describing light diffusion inside bounded media, and by study of the angular dependence of diffusely transmitted light it has been accurately measured for various interfaces. For our particular case of air–glass–water interfaces, its value is 1.77.[14] It is worth mentioning that the $s^{-5/2}$ behavior of the energy flux corresponds to diffusive waves with large optical path lengths.

The path-length-resolved backscattered intensities that correspond to water suspensions of polystyrene microspheres with particle diameters of 0.426 μm and various volume fractions are shown in Fig. 1. The data are normalized with the intensity of the incident field. For clarity, the values of normalized backscattered intensities have been scaled as indicated. To account for instrumentation effects we use a twofold fitting procedure. First, the trail of the distribution is fitted with $-5/2$ dependence to yield an amplitude normalization constant and, second, the whole distribution is fitted with the prediction of Eq. (1) to infer the value of l_t. In the present experiments, absorption effects have been neglected because the absorption lengths of the media are roughly 2 orders of magnitude longer than the corresponding scattering lengths. As we can see from Fig. 1, the diffusion theory, corrected for the effective reflectivity of the interface, makes a good description of the experimental data that correspond to various volume fractions. Applying the Mie scattering theory, we also calculated the corresponding values of the transport mean free paths for these media. Excellent agreement was obtained between the conventional estimations of l_t and the results of our fitting procedure. For samples with volume fractions of 2.5%, 5%, and 10%, the measured l_t values of 197, 101, and 49.2 μm are to be compared with, respectively, 206, 103, and 51.5 μm obtained from Mie theory. These results suggest that reliable measurements of photon transport mean free path can be based on OPS.

Finally, we comment on the potential of OPS as an investigation method and mention some of the areas

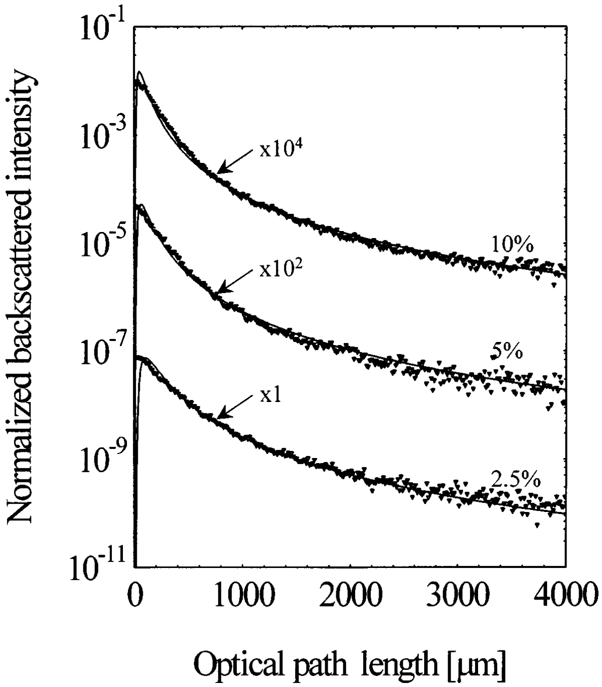

Fig. 1. Values of normalized backscattered intensity corresponding to suspensions of polystyrene microspheres with diameters of 0.426 μm and with volume fractions of 10%, 5%, and 2.5%. Continuous curves, the fit with the functional form in Eq. (1). For clarity, the normalized intensities have been scaled as indicated.

for which this new technique can be relevant. One can directly obtain the optical path-length probability density $P(s)$ by normalizing the low-coherence reflectivity data; subsequently, one can infer characteristics such as the average optical path length as well as higher-order moments for any kind of random medium. A detailed description of field penetration depth is important in many light-delivery applications and becomes crucial for medical techniques such as photodynamic therapy. For brevity, in this Letter we have neglected absorption, but incorporating its effects is straightforward. It is well known that the correlation function measured in diffusive wave spectroscopy is merely the Laplace transform of $P(s)$,[15] and we anticipate that a direct experimental evaluation of $P(s)$ will permit a more accurate analysis of diffusive wave spectroscopy data. As we mentioned above, the parameter z_0/l_t is intimately related to the boundary reflection effects on the wave transport within the medium. Although this parameter can be calculated when the refractive indices at the interface are known, experimental access to it for arbitrary samples has been limited, so far, to angularly resolved diffuse transmission.[14] OPS provides a high-resolution $P(s)$ distribution, and it can be used for accurate measurements of the boundary reflection properties of unknown systems.

In conclusion, we have introduced a new approach to measuring the optical path-length distribution of light scattered from a random medium. The proposed optical path-length spectroscopy is not limited to the study of dynamic randomness or to backscattering geometries. The results presented in this Letter show that accurate data about the transport mean free path can be obtained and that the less-demanding instrumentation needed for recording OPS data makes this approach appealing for a variety of applications.

This research was partially supported by the National Science Foundation under grant ERC-94-02989 and by the University of Central Florida through its Florida High-Tech Corridor initiative. A. Dogariu's e-mail address is acd@creol.ucf.edu.

References

1. R. C. Youngquist, S. Carr, and D. E. Davies, Opt. Lett. **12,** 158 (1987).
2. J. M. Schmitt and S. H. Xiang, Opt. Lett. **23,** 13 (1998).
3. D. A. Boas, K. K. Bizheva, and A. M. Siegel, Opt. Lett. **23,** 5 (1998).
4. A. Ishimaru, *Wave Propagation and Scattering in Random Media* (Academic, New York, 1971), Vol. 1, Chap. 9.
5. A. Ishimaru, J. Opt. Soc. Am. **68,** 8 (1978).
6. M. S. Patterson, B. Chance, and B. C. Wilson, Appl. Opt. **28,** 12 (1989).
7. K. M. Yoo, F. Liu, and R. R. Alfano, Phys. Rev. Lett. **64,** 22 (1990).
8. F. Liu, K. M. Yoo, and R. R. Alfano, Opt. Lett. **18,** 6 (1993).
9. A. Dogariu, C. Kutsche, P. Likamwa, G. Boreman, and B. Moudgil, Opt. Lett. **22,** 9 (1997).
10. D. J. Durian, Phys. Rev. E **50,** 5 (1994).
11. P. A. Lemieux, M. U. Vera, and D. J. Durian, Phys. Rev. E **57,** 4 (1998).
12. H. Chou and W. V. Sorin, Hewlett-Packard J. **44**(1), 52 (1993).
13. J. J. Duderstadt and L. J. Hamilton, *Nuclear Reactor Analysis* (Wiley, New York, 1976).
14. M. U. Vera and D. J. Durian, Phys. Rev. E **53,** 4 (1996).
15. D. J. Pine, D. A. Weitz, J. X. Zhu, and E. Herbolzheimer, J. Phys. (France) **51,** 2101 (1990).