Measurement of the anomalous phase velocity of ballistic light in a random medium by use of a novel interferometer

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Ballistic light, i.e., radiation that propagates undeflected through a turbid medium, undergoes a small change in phase velocity and exhibits unusual dispersion because of its wave nature. We use a novel highly sensitive differential phase optical interferometer to study these previously unmeasurable phenomena. We find that ballistic propagation can be classified into three regimes based on the wavelength-to-size ratio. In the regime in which the scatterer size is comparable with the wavelength, there is an anomalous phase-velocity increase as a result of adding scatterers of higher refractive index. We also observe an anomaly in the relative phase velocity, where red light is slowed more than blue light even though the added scatterers are made of material with normal dispersion. © 2001 Optical Society of America

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Ballistic light is defined as light that traverses a scattering medium in the same direction as incident light.[1] Ballistic light (along with its acoustic analog) is particularly important for biomedical imaging applications.[2,3] Conventionally, ballistic propagation is pictured as photons that are undeflected in transmission. This picture, henceforth called the photonic model, is used extensively in optical tomography,[2] and it adequately explains many properties of ballistic propagation. However, this model is incomplete, as the wave nature of light is not considered. In this Letter we study the phase-velocity and -dispersion characteristics of ballistic light waves traversing a diffuse scattering medium as a function of scatterer size. To study these characteristics optically, we use a novel interferometer capable of measuring very small differences in phase velocity between the wavelengths 800 and 400 nm. This interferometer is sensitive to phase-velocity differences of 40 m/s in a 2-cm-thick turbid sample. This remarkable sensitivity permits us to study dilute turbid media—a relevant model for optical applications such as biomedical imaging[4] and remote sensing through fog.[5]

We show that our experimental data agree well with the predictions of the van de Hulst and Mie scattering theories.[6] Both the theory and the experiment demonstrate that ballistic propagation separates into three regimes: (1) When the scatterer size (a) is much smaller than the optical wavelength (λ), the turbid medium can be approximated as a bulk medium for phase-velocity considerations; (2) when a is comparable to λ, the phase velocity is strongly dependent on scatterer size; (3) when a is much larger than λ, turbidity can be ignored for phase-velocity considerations. In the second regime, ballistic light can propagate with a phase velocity that is uncharacteristic of the constituent materials. Hence the ballistic light itself must carry phase information about the structure and composition of the turbid medium. The photonic model simply cannot explain this variation in phase velocity. We note that this effect is very small (approximately 1 part in 10^6) in a typical diffuse turbid medium and thus, to our knowledge, has remained unobserved until now.

We observe this phenomenon by using a novel low-coherence phase-dispersion interferometer (Fig. 1).[7] The input light is created by superposing beams of laser light at the fundamental and the second-harmonic frequencies. The source is a low-coherence Ti:sapphire laser producing 150-fs pulses at 800 nm, and the second harmonic is generated by a standard frequency doubler. The superposed beam is split into two components at a beam splitter. One component makes two passes through the turbid medium in the signal arm of the interferometer. The other component passes through a compensator cuvette of water and reflects from a reference mirror in the reference arm. The reference mirror moves uniformly at a speed of 1 mm/s and induces a Doppler shift in the return beam. The recombined beams are then separated by wavelength by use of a dichroic mirror and are detected separately. The resulting heterodyne signals at both wavelengths are measured and digitized by a 16-bit 100-kHz analog–digital converter. Each digitized signal is bandpassed around its center heterodyne frequency, which is given by the Doppler shift. The filtered signals are then Hilbert

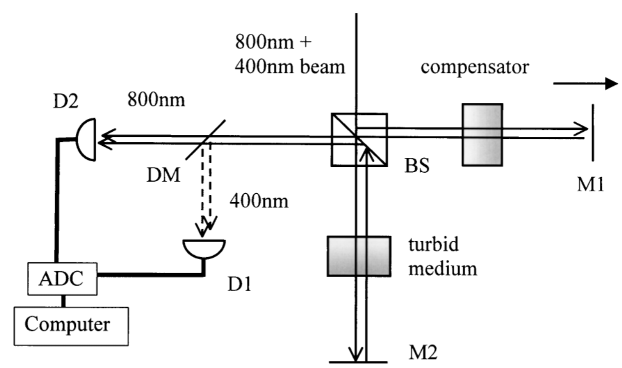

Fig. 1. Experimental setup: M1, M2, mirrors (M1 is the reference mirror); BS, beam splitter; D1, D2, photodetectors; DM, 400-nm–800-nm dichroic mirror; ADC, analog–digital converter.

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transformed, and the phases, Ψ_1 (fundamental) and Ψ_2 (second-harmonic), are extracted.[8,9] Related phase techniques have been used to measure the dispersion of metals,[10] the refractive index of air,[11] and plasma electron density.[12]

Jitter in the path lengths of the interferometer arms due to, for example, vibrations of the mirrors will introduce correlated phase errors between the heterodyne signals. This novel interferometric design eliminates these phase errors through comparison of the two heterodyne phases to achieve sensitivity that is unattainable by conventional means. Specifically, we substract twice Ψ_1 from Ψ_2 to obtain the difference in the optical path lengths of the two wavelengths, $\Delta L_{k_2,k_1}$, with great sensitivity (~5 nm; Ref. 7):

$$\Delta L_{k_2,k_1} = \frac{\Psi_2 - 2\Psi_1}{k_2}\,. \tag{1}$$

The experiments measure the phase of light traversing a 10-mm-thick turbid medium composed of scattering polystyrene spheres in water. A water-filled cuvette of the same thickness provides phase compensation. Measurements are taken as polystyrene microspheres of a given size are gradually added to the signal-arm cuvette. The fractional volume of microspheres, η, is varied from 8×10^{-6} to 3×10^{-3}. The relative refractive index of the microspheres with respect to that of water is 1.20 at 800 nm and 1.23 at 400 nm. Each measurement of the optical path difference is then used to find the fractional phase-velocity difference, $\Delta v_2/v_0 - \Delta v_1/v_0$, between the two wavelengths in the cuvette:

$$\frac{\Delta v_2}{v_0} - \frac{\Delta v_1}{v_0} = -\frac{\Delta L_{k_2,k_1}}{n_0 L}\,, \tag{2}$$

where v_0 is the speed of light in water and n_0 is the refractive index of water. Note that the insignificantly small second-order corrections that are due to dispersion of water are omitted. Measurements are made for a succession of microspheres varying in radius from 10 nm to 10 μm. The data points in Fig. 2 show the measured fractional difference in phase velocities as a function of scatterer size. The experimental observations are best discussed by comparison with the following theoretical phase-velocity analysis of the ballistic light.

The transmission of ballistic light through a turbid medium can be characterized by a complex relative refractive index, $n_{\mathrm{cx}} = n - in'$. The ballistic light field, $E(L)$, which has traversed a distance L in the turbid medium, can be written as a complex exponential attenuation of the incident field, $E(0)$:

$$\begin{aligned} E(L) &= E(0)\exp(-ikn_{\mathrm{cx}}L) \\ &= E(0)\exp[-ik(n - in')L]\,, \end{aligned} \tag{3}$$

where k is the wave number. n_{cx} can be expressed in terms of $S(0)$, the scattering function evaluated in the exact forward direction of the input light (Ref. 4, p. 33):

$$n = 1 + \frac{2\pi N}{k^3}\ \mathrm{Im}[S(0)]\,, \tag{4a}$$

$$n' = \frac{2\pi N}{k^3}\ \mathrm{Re}[S(0)]\,, \tag{4b}$$

where N is the number of scatterers per unit volume.

The imaginary part of the refractive index is associated with the well-known attenuation of ballistic light that is due to scattering and has been studied extensively.[2] However, the effect of scatterers on the real part of the refractive index is small and cannot be readily measured by conventional means. Our highly sensitive interferometer allows us to study the subtle variations of the imaginary part of the scattering function.

To elucidate the effect of spherical scatterers on the refractive index (or, equivalently, the associated phase velocity), we consider the van de Hulst scattering model for spheres of radius a and relative refractive index m (Ref. 6, pp. 174–179). This model is strictly valid only when the scatterer size is large compared with the wavelength and the refractive-index difference is small. Nevertheless, the model provides important physical insights into the scattering process and, as shown below, describes the salient features of ballistic light propagation well beyond these limits. For light at one wavelength, the van de Hulst model gives a fractional phase-velocity change of the form

$$\frac{\Delta v}{v_0} = 1 - n = -\frac{3\eta}{2a^3k^3}\,(ka)^2\left(\frac{\sin\rho}{\rho^2} - \frac{\cos\rho}{\rho}\right), \tag{5}$$

where $\rho = 2ka(m - 1)$ is the normalized scatterer size and $(m - 1)$ is the relative refractive-index difference between the scatterers and the surrounding medium. A plot of $\Delta v/\eta v_0$ by use of the van de Hulst model is shown in Fig. 3. For comparison, an exact computation based on Mie theory[6] is also shown.

Figure 3 reveals three different regimes of ballistic light propagation, as discussed above, depending on the scatterer properties. We can discuss each of these analytically, using the van de Hulst model:

(1) $\rho \ll 1$—*turbid medium as bulk medium.* In this limit, the phase-velocity change, Δv, reduces

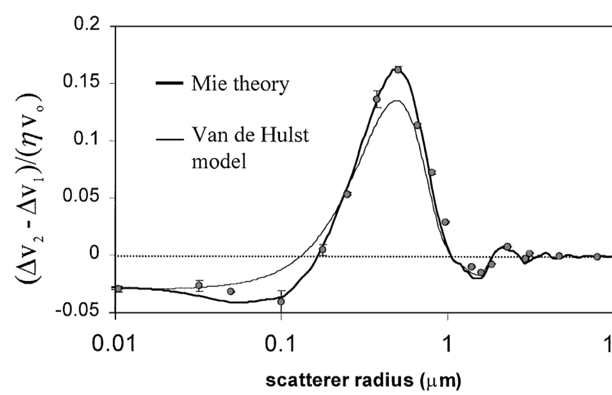

Fig. 2. Phase-velocity difference versus scatterer radius. The data points show the measured values. Theoretical fits based on the van de Hulst model and the Mie theory solution are shown.

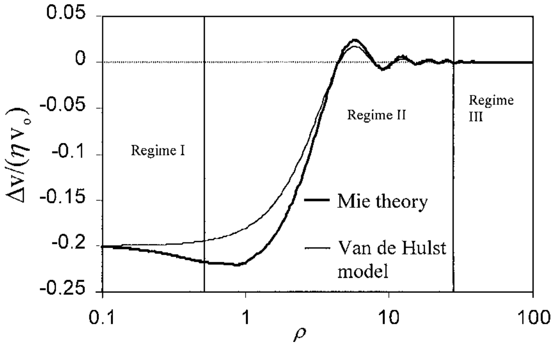

Fig. 3. Modeled normalized phase velocity $\Delta v/\eta v_0$ versus normalized scatterer size ρ. The normalized refractive-index difference, $(m - 1)$, equals 0.2 for this plot.

to $-\eta v_0(m - 1)$. This change arises only from bulk refractive-index change that is due to the presence of small scatterers. From another perspective, when the phase lag through each scatterer is small, the net result is simply an overall change in phase velocity, as determined by the refractive-index difference.

(2) $\rho \approx 1$—*no simplification.* In this regime Eq. (5) cannot be simplified. The phase velocity is seen to oscillate with changing ρ. The net change in phase velocity is strongly dependent on whether the forward-scattered light is in or out of phase with the input light. We note the existence of an anomalous phase-velocity increase for some values of ρ, despite the fact that the scatterers have a higher refractive index than water.

(3) $\rho >> 1$—*phase velocity is independent of turbidity.* In this limit, Δv is zero. This is the only regime for which the photonic model provides a complete description. The phase velocity is thus independent of the presence of turbidity. Physically, we can understand this from the fact that when ρ is large the phase of the transmitted light varies rapidly with increasing distance from the center of the sphere. The net result is that the phase shift of the transmitted light averages to zero. Therefore large scatterers have no effect on the bulk refractive index for ballistic propagation.

The discussion presented above is based on the behavior of ballistic propagation for light of a single wavelength. In our experiments, based on phase-velocity differences between two wavelengths, the three regimes still can be clearly seen (Fig. 2).

The change in phase velocity at 400 nm compared with that at 800 nm reveals another anomaly. Usually, adding a material with an index higher at 400 than at 800 nm (normal dispersion) to water would cause the 400-nm light to be slowed more than at 800 nm. However, in the case of scatterers with $\rho \approx 1$, the opposite is observed. This effect is due to the shift in the phase-velocity profile that arises from the scaling of ρ with wavelength (Fig. 3) and is not dependent on the anomalous phase-velocity increase discussed above.

The distinctive features of the phase-velocity difference profile should make it possible to extract precise scatterer-size distributions in polydisperse media by scanning of the fundamental–second-harmonic wavelengths. High precision is afforded by the extremely high sensitivity achieved with phase-based measurements. This method should complement related intensity-based techniques for measuring the size distribution of cell nuclei, an important indicator of precancerous changes in biological tissues.[13]

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