Multiple Scattering Probes of Disordered Materials

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Introduction

A large class of materials of great technological importance and scientific interest are highly disordered on length scales comparable to, or larger than, the wavelength of light. As a consequence of this disorder, these materials are often composed of a large number of replicas with different indices of refraction, separated by interfaces that are abrupt on the scale of the wavelength of light. Each of these interfaces refracts or scatters light, making these materials quite opaque optically. Even in the absence of absorption, the strong scattering precludes the direct transmission of light rays through the material. This greatly complicates the study of these materials.

Examples of these disordered materials abound. Many are complex fluids; colloidal suspensions and other dispersions such as foams and emulsions often consist of particles or droplets whose diameters are comparable to the wavelength of light. Unless they are very dilute, these dispersions are typically highly opaque. Other materials consist of solid grains of similar sizes without the surrounding fluid. For example, most ceramics are composed of oxide grains sintered together; the voids between the grains lead to the large number of interfaces, and hence to the strong scattering and opaque appearance commonly associated with these materials.

Similarly, many nonequilibrium states also possess a high density of voids that scatter light; filters, oxide catalyst supports, and even rocks are all optically opaque because of the strong scattering. Sand and other unconsolidated granular materials are also opaque in part because of the large number of interfaces that scatter light. Finally, there are many important bio-

Figure 1: Schematic representation of the random diffuse photon paths that result in the diffuse transmittance of light through a multiply scattering sample. The light intensity decreases through the sample, and each path represents a random walk that a photon follows in traversing the sample.

terials that scatter light so strongly as to preclude the use of optical probes of their properties; the most important of these is the human body. These examples of disordered materials that are optically opaque due to the large number of interfaces are representative, but far from all-inclusive.

The first step in the study of any material is to determine its structure; this is followed by a study of its properties. Because the size of the essential structures in this class of disordered materials is comparable to the wavelength of light, optical probes are ideally suited for studying both their structure and their properties; however, precisely because the structures are comparable to the wavelength of light, and thus lead to such strong scattering, the use of optical probes is precluded. Instead, rather specialized techniques must be employed to probe these materials. For example, many materials are dispersions of either solids or droplets of liquids immersed in a fluid; for these dispersions, it is sometimes possible to judiciously choose the fluid to exactly match the indices of refraction of all the components to minimize the scattering and allow direct visualization of the material. Alternatively, modern x-ray tomography techniques can be used to study the structure of small samples of some of these disordered materials; x-rays are prone to strong absorption, but do not scatter as strongly as light. However, despite the advantages of these direct imaging methods, their application is limited.

Despite the limitations imposed by the very strong scattering in these materials, optical techniques still possess great potential for studying the properties of these materials. Fortunately, there has been considerable progress in the development of several different optical probes of strongly scattering media. These methods are all based on the recognition that the propagation of light through very strongly scattering media can be well described using the diffusion approximation. This allows the propagation of the light to be described in a simple, statistical fashion. The essential approximation used in the diffusive propagation approximation is that the phase of all the scattered light is so randomized that any interference effects within the medium can be neglected. As a result, only the intensity of the light need be considered in the description of the propagation of the light. This intensity propagation can be very well approximated as a diffusive process. Then, physically, the paths followed by the photons propagating through the medium can be described as diffusion, and hence follow random walks. A physical representation of this is illustrated in Figure 1, where some of the diffusive paths followed by the photons propagating through a strongly scattering medium are illustrated.

The statistics of these random walks are well understood; both the distribution and the length of these paths can be determined through the solution of the diffusion equation for the light. However, since only the diffusing intensity is relevant, each of these diffusive photon paths is statistically uncorrelated with all other paths except with the path itself. Nevertheless, each path still contains critical phase information; recognition of this fact provided the critical insight that underlies many recent advances. This phase infor-
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mation reflects the length of the optical path that the diffusing photons have fol-
lowed on traversing the medium. It can be determined by the arrival time of the
photons if they are incident as a short pulse. Alternatively, it can be determined
through scattered light, allowing the study of either an independent reference beam,
the light from other, independent diffus-
ion paths. Furthermore, the relatively small fraction of the light in-
tensity that traverses a very strongly scat-
tering medium, the light photon fluxes easily available with modern lasers, com-
pared with the very high sensitivity of optical detectors, make it possible to mea-
sure this phase information. This has led to the development of significantly new optical
probes of strongly scattering materials.

The goal of this article is to review some of these new methods and to provide
some perspective on their utility and their applications. In particular, we consider
two distinct classes of techniques that take two divergent approaches. In the first
approach, the consequences of the multiple
scattering of the light are minimized.
This is typically achieved through time
resolution; light which has been more
strongly scattered follows a longer path,
thereby taking a longer time to traverse
the medium. By contrast, light that is un-
scattered, or scattered only a few times, traverses the medium much faster. Thus,
by time resolving the transmitted light, it becomes possible to determine the
time the light is to that which is either unscattered or only
very weakly scattered allowing the direct imaging of even very opaque materials.
In the second approach, the strongly
scattered light is measured. When
scattering approximation is used to describe
its properties, the small fraction of light that is
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In the second approach, the strongly
scattered light is measured. When
scattering approximation is used to describe
its properties, the small fraction of light that is
scattered can be directly measured by observing the change in the intensity of the
transmitted light. In this approach, the
transmission coefficient measured in this
way depends on the size and shape of the
particles in the medium. These systems,
where the scatterers are essen-
tially a distribution of particles, the trans-
mittance coefficient depends on the size,
structure, density, and dielectric constant of
the scatterers as well as on the spatial
variations in the dielectric constant of the
medium. These variations in effective index
are frequently associated with sharp inter-
facial boundaries, such as those occurring in
biological tissues, emulsions, colloids, or
tissues. These systems, where the scatterers are essen-
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the scatterers as well as on the spatial
correlations between them. It should be possible, therefore, to extract useful infor-
mation about a material by measuring its
transmission coefficient. Varying the optical wavelength changes the length
scale of the probe and may provide addi-
tional information. To make contact be-
tween optical measurements and material
properties, one must be able to describe the transport of light through a multiply
scattering sample.

The diffusion approximation is the most
common approach for describing light
transport through multiply scattering me-
dia. In this approach, a process traversing a
sample execute trajectories which are de-
scribed statistically as random walks, as il-
lustrated schematically in Figure 1. The
only important parameter is describing the
courtyard size, I, commonly called the transport mean free path. The diffusion approximation
relies on the transport mean free path to the mea-
sured transmission coefficient: T = e-2l,
where I is the sample thickness. The
goal of any useful theory, then, is to relate
T and the structural properties of the mate-
rials under study.

Within the diffusion approximation, one
assumes that each photon scatters many
times and that the total transmitted light intensity is given by the incoherent sum of intensities for all possible photon paths through the sample. The distance between consecutive scattering events is the scat-
tering mean free path, l, if the number density, n, of scatterers is small,

$$I = l = \frac{\mu n}{\mu_s} \mu$$

where n is the total scattering cross sec-
tion. The mean free path of a photon is not rarer
than the transport mean free path. This is because materials that multiply scatter
light typically consist of particles of (or mi-
crostructures) whose size is comparable to a light's wavelength. Even smaller (smaller particles usually do not scatter strongly enough to cause significant mul-
tiple scattering). These particles scatter more strongly in the forward direction so that the photon's path is not ran-
domized by a single scattering event; the random walk length, $l$, is less than l. The
transport mean free path or random walk step size is related to the scattering mean free path by

$$l = \frac{1}{n} \text{cos} \theta$$

where $\theta$ is the average cosine of the scattering angle for single scattering. Ex-
pressions similar to Equation (2) appear in equations for the persistence length of
polymers and the resistivity of metals.
If there are changes in the size, shape, or
spatial distribution of scatterers, $\theta$ will change. If the number density of scat-
ters changes, $I$ will change. Equations (1)

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and (2) provide us with the starting point we need to relate changes in \( p \) to changes in the structure of a sample under investigation. We now turn to some examples of how these ideas have been applied to a few different systems.

One particularly simple but elegant application of these ideas has been the use of transmission measurements to follow the time evolution of a negatively stained system, namely, living cells. In this experiment, the transmission coefficient of light through slicing was observed to decrease in the square root of time, indicating that \( p \) was increasing approximately as the square root of the number of particles of size \( k \) present in the sample. By adjusting \( k \), one can move through the maximum and minimum in \( p \).

Another exciting recent development has been the use of multiple scattered light for imaging purposes. The goal is to detect objects and structures hidden from direct visual observation inside of optically turbid media. The motivation for much of this work has been medical imaging applications, and a variety of schemes have been developed. A particularly simple scheme conceptually is to launch a very short (microsecond) pulse of light into a sample and to use time gating to detect only the light that is transmitted through the sample without being scattered. The idea is that the propagation delay time for unscattered light, \( t \), where \( c \) is the speed of light in the sample, is much shorter than the propagation delay time for multiply-scattered light, approximately \( t \propto V_p \). This unscattered light arises as a factor of \( t \) and \( p \) (typically \( p \gg 1 \)) at positions of the intersection of the direct light path with the sample. Experiments have shown that essential parts of the sample can be imaged from the intersection of the unscattered light that is, from light which is scattered within only a small forward-angle scattered light cone so that their paths are essentially ballistic. The advantage, of course, is larger signal levels. A drawback of these gating schemes is that they require costly equipment both to excise the pulse and to detect light with adequate temporal resolution. Moreover, the intensity of unscattered light is attenuated approximately as a factor of \( 1 - exp(-\alpha t) \) which is typically \( 10^{-3} \) to \( 10^{-4} \). Nevertheless, this technique has been successfully demonstrated in model systems with attenuation factors of \( 10^4 \). The reason such high attenuation factors can be tolerated is the high intensity available from modern lasers and the sensitivity of light detectors.

Other imaging schemes utilize rather than discard the highly multiply scattered light. The advantages are a much larger signal and less costly equipment. These advantages are partially offset by decreased spatial resolutions. One particularly promising scheme involves using amplitude modulated (AM) continuous-wave laser sources. The idea is that light whose intensity is modulated at an angular frequency \( \omega \) and which diffuses with a diffusion coefficient \( D = \omega / 2 \pi \) through a turbid medium consists of crests and troughs in the light intensity. These crests and troughs are spatially separated by approximately \( \Omega = 1.25 \) the mirror displacement of a photon in the modulation period \( 2 \pi / \Omega \). The crests and troughs can be viewed as a damped propagating wave with wavelength of \( 2 \pi / \Omega \).

Another aspect of light scattering that has long been a very useful probe of the dynamics of systems that possess structures with length scales comparable to the wavelength of light. The most common technique is dynamic light scattering (DLS), which entail an analysis of the temporal fluctuations of the scattered intensity. This technique exploits the coherence of the scattered light; if the incident light is spatially and temporally coherent across the sample volume, the scattered intensity will show a spatially random pattern of intensity maxima, called speckle spots. These speckles represent the diffraction pattern of the light from the whole sample; their size is determined by the angle subtended by the sample at the detector times the wavelength of the light. The intensity in each speckle spot will fluctuate randomly in time as the sample changes. The characteristic scale of these fluctuations is determined by the wavelength of light. Each speckle spot results from the interference of all the light scattered from the sample; therefore the intensity of the speckle spots will change significantly when the relative phase of the light scattered from each point in the sample changes by \( 2 \pi \). This will occur when each scattering region in the sample moves by roughly \( q \), where \( q \) is the scattering vector. Since \( q \propto l - x \), DLS is sensitive to motion or dynamics on a length scale on the order of the wavelength of light.
single specie spot. To obtain meaningful data from the random temporal fluctuations, the intensity autocorrelation function is measured. Because the temporal fluctuations are random, the intensity autocorrelation is a decaying function of time difference, and DLS measurements yield the characteristic decay time, $\tau$. To analyze these data, the measured decay time must be related to the dynamics of the scattering system. In general, this involves determining the temporal evolution of the motion of the scatterers; $\tau$ is the time required for a typical scatterer to move by $a$. For example, in a simple case of a dilute suspension of small particles undergoing Brownian diffusion, the characteristic decay time is $\tau = 1/(4D)$, where $D$ is the diffusion coefficient of the particles. Thus, to relate the time scale measured experimentally to the motion of the scatterers requires knowledge of a length scale, which is set by $a$. This scattering wave vector is only well-defined in the limit of single scattering, restricting the application of traditional DLS and precluding its use for the study of opaque materials.

While traditional DLS cannot be used to study the dynamics of strongly scattering disordered materials, some DLS techniques can still be employed. If a strongly scattering medium is illuminated with coherent light, the multiply scattered light still contains information about the path length due to the dynamics of the scatterers. Moreover, the temporal autocorrelation of the intensity fluctuations still decays with a characteristic time. The challenge is to relate this characteristic decay time to the dynamics of the medium. This can be done, allowing the motion to be determined about the dynamics of the scattering medium. The resultant light-scattering technique is called diffuse wave spectroscopy (DWS).$^{13,14}$ Not only does DWS allow the study of disordered materials that strongly scatter the light, but it specifically exploits the multiple scattering of the light to allow the study of new regimes of dynamics that would not otherwise be accessible.

The central assumption that is made in the analysis of DWS is again that the propagation of the light through the scattering medium can be described by the diffusion approximation. This allows the distribution of diffusing photon paths, and their lengths, to be determined. Moreover, since the contour lengths of the paths are known, the number of scattering events that the photon undergoes can also be determined, provided the transport mean free path, or photon randomization length, is known. The dynamics of the scattering medium will cause the scatterers to move, and the length of each path will change. This corresponds to a change in the relative phases of the paths, resulting in temporal fluctuations of the scattered intensity through the interference with light from other paths. Because the paths are uncorrelated, the total autocorrelation function of the temporal fluctuations of the scattered intensity is the sum of the contributions from all the individual paths. However, since the intensity in each specie spot still reflects the interference of light from the different paths, it will change when the relative phases of the paths vary by $2\pi$, or when the changes in the total length of the paths is on the order of a wavelength of light. This is a critical difference between traditional dynamic light scattering and diffusing wave spectroscopy; in DLS, the dynamics result from the motion of a single scatterer over a distance on the order of a wavelength of light, while in DWS, the dynamics result from the change in the length of a diffusive light path on the order of a wavelength.

Within these approximations, the determination of the total autocorrelation function requires the calculation of the contribution of each of the uncorrelated diffusive light paths. The change in the phase of the total path will reflect the aggregate change in path length due to the motion of all the scatterers that comprise the path. This can be determined by analyzing a statistical approach to isolate the correlation function of the total path in the presence of an average or typical scattering event. Since both the contour length of the path and the scattering length are known, the total number of scattering events that comprise the path can be determined. A long path consists of a large number of scattering events, each of which is independent. Thus, to a first approximation, the autocorrelation function of the total path will be the product of all the independent correlation functions of the individual scattering events. Moreover, since the total correlation function consists of the contributions of a large number of scattering events, this provides a very effective ensemble average, allowing each of the individual scattering events to be replaced by a single average scattering event, where the averaging reflects the scattering probability of the medium. Then, the total correlation function will be the sum of the individual contributions of all the paths of different lengths, weighted by the probability that the diffusing photons follow that path. Following this scheme, the total autocorrelation function of the scattered intensity fluctuations can be calculated.

The correlation function of the multiply scattered light measured in a DWS experiment reflects the dynamics of the scattering medium. It involves many of the same quantities as does a correlation function measured in a traditional DLS experiment. The time scale still involves the dynamics of the scatterers, and their motion over the length scale of a wavelength. However, a DWS experiment is sensitive to the aggregate motion of a large number of scatterers that make up each path. Thus, DWS scatterers need move a much smaller distance to make the total path length change by a wavelength. As a consequence, a DWS measurement is sensitive to motion over much smaller length scales than a traditional DLS measurement. The length scale of the motion that is probed by DWS depends on the total length of the scattering paths that contribute, and the number of scattering events in each. For example, if the light is transmitted through a slab of thickness $L$, the average number of randomization events in the characteristic path is on the order of $L^2/a^2$. The time scale for the decay of this path is decreased by the inverse of this number; the length scale over which a scatterer must move is decreased by a corresponding amount. This is a primary advantage of DWS; it probes the dynamics of the scattering medium over length scales that are significantly shorter than those probed by traditional DLS, where the light is multiply scattered, some information is lost. It is not possible to probe motion on the order of a typical scattering event in DLS. Instead, DWS probes an event averaged over many events. Moreover, because the diffusion approximation is used to interpret the results, knowledge of $a$ is essential to interpret the results. However, $a$ can be determined independently by independent measurements of the total transmission.

Diffusing wave spectroscopy experiments are performed in a fashion very similar to the traditional DLS experiments. The scattered light within a single correlation time is detected and the temporal autocorrelation function of the intensity fluctuations is measured. However, since the light is strongly multiply scattered, it is 40% of size in all directions. Therefore, unlike DLS, the scattering angle is not well-defined. Instead, there are only two useful geometries for DWS experiments, either transmission or backscattering. The reason for this is that the two geometries are the distribution of diffusive photon paths that contribute to the auto-
correlation functions. In backscattering, long paths can contribute; however, a significant contribution arises from very short paths, where the light is scattered only a few times and exits the sample from the same side it entered. While the functional form that describes the autocorrelation functions in this case is very simple, the contribution of the very short path can not be easily described within a diffusion approximation, since the length of these paths is comparable to the transport mean free path, the minimum distance over which a diffusion approach can possibly apply. This complicates the interpretation of the results significantly; as a result, the backscattering geometry has found relatively fewer applications. By contrast, in transmission, the light must traverse through the whole sample; as a result, all of the paths can be well described by the diffusion approximation. There is a characteristic length scale for the paths, L_D, which can be experimentally controlled by varying the sample thickness. Thus, the exact form of the autocorrelation functions measured by DWS depends on the geometry of the experiment.

To illustrate the unique features of DWS, we consider a concentrated suspension of uniformly sized colloidal particles. In this case, the particle motion is simply Brownian diffusion, and the functional forms for the DWS autocorrelation function can be inverted to determine the root-mean-square (rms) displacement of a particle as a function of time. As an example, we use DWS to probe a suspension of 1.53 μm diameter polystyrene latex spheres, suspended in water at a volume fraction of ~0.02. Motion on length scales as short as a few angstroms can be observed.

fluid, the wake generated by the motion of the particles causes their velocity correlation function to decay algebraically rather than exponentially, leading to the very slow approach to the asymptotic diffusive form. These data clearly exhibit the very high resolution of DWS, and its unique ability to probe motion over very short length scales. In fact, the resolution illustrated by this example is not the inherent limitation of DWS, but by using a thicker sample, motion over even shorter length scales can be measured. This was done by Zhao and collaborators, who used a Michelson interferometer to measure correlations over even shorter time scales. Using thicker samples, they were able to achieve resolutions on the subangstrom level.

Diffusing wave spectroscopy is ideally suited for applications involving very concentrated suspensions. These are exactly the sort of systems that cannot be studied with traditional DLS because of the problems of multiple scattering; yet these are also the colloid systems of greatest technological importance. Perhaps one of the most important applications of DWS is for measuring the size and size evolution of colloidal particles in concentrated suspensions without the requirement of dilution needed for traditional DLS. For example, Hovsepian has studied the culling of milk, which is the precursor to making cheese. In this process, the emulsion droplets of fat that comprise the milk aggregate together and ultimately form a gel network. Using DWS, Horne was able to follow this aggregation and subsequent gelation. These experiments were performed primarily in the backscattering geometry. Other developments of DWS for particle sizing have emphasized the transmission geometry. Van Keuren and collaborators used two single-mode optical fibers immersed directly into the suspension, one for the source and the second for the detector. This provides a simpler functional form for the DWS autocorrelation function, and allows particle sizing measurements to be performed more directly. With continued development, DWS could potentially become an important technique for process monitoring in the manufacture of colloidal particles. Diffusing wave spectroscopy has also played a role in the study of several important problems in colloid physics. For example, Chatkin and his collaborators have studied the behavior of fluidized beds using DWS. These are suspensions of relatively large colloidal particles; gravitational settling is counteracted by the upward flow of the suspending fluid. Since the particles are undergoing flow, DWS probes the relative velocity differences, or

![Figure 2. Root-mean-square displacement of a particle as measured with diffusing-wave spectroscopy. The particles are 1.53 μm diameter polystyrene latex spheres, suspended in water at a volume fraction of ~0.02. Motion on length scales as short as a few angstroms can be observed.](image-url)
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the shear. In a fluidized bed, the relative velocities are random, so DWS probes the variance in the particle velocities. Chaikin and his collaborators used DWS to measure this variance as the flow rate was varied, and hence the volume fractions of the fluidized bed was changed. The variance was found to decrease with volume fraction, approaching zero near a volume fraction of about 0.4, significantly lower than the volume fractions where the average velocity approaches zero. While DWS is ideally suited to measuring some of the important quantities in fluidized beds, it may also be used to study the behavior of fluidized beds prepared with granular particles and fluidized by the flow of a gas. The behavior of granular systems such as this is addressed in another article in this issue. The physics of these systems is not well understood, and the application of DWS may help address some of the key problems.

Another very elegant application of DWS is in the study of electrochemical (ER) suspensions by Ginder. Electrochemical suspensions are dispersions of colloidal particles that are subjected to an electric field, which induces dipole moments in the particles. If the dipole interactions are strong enough, the particles attract one another, forming chainlike structures that can transport charge when a current is applied to the field. These chains can modify the viscosity of the suspension, turning a fluid mixture into a solid-like paste. This feature makes ER fluids potentially attractive for a variety of applications, such as switches, provided that the response time of the ER fluid of interest is fast enough. Ginder used multiple light scattering techniques to probe the response of an ER fluid to an electric field. For example, Ginder found that he could measure the response time of an ER fluid to the sudden application of an electric field by monitoring the increase in intensity of multiply scattered light as the ER fluid is passed through the sample. The increase in transmitted intensity occurs because of the increase in local order between particles induced by the electric field. Ginder also used DWS to monitor the motion of the particles in ER fluids in response to small oscillating electric fields. These fields were kept small enough so as to not change the transmission coefficient of the multiply scattered light; the oscillations were observed to the measured autocorrelation functions arise from temporal phase fluctuations produced by the electrodynamic motion of the suspended particles making up the ER fluid. Ginder concluded from the DWS measurements that electrophoresis does occur in these systems and that it can promote particle aggregation at high applied electric fields.

Although many of the applications of DWS to date have entailed problems involving colloid suspensions, the measured autocorrelation function will reflect the dynamics of the sample studied. An example of the use of DWS to study a completely different form of dynamics that also highlights another unique feature of the technique is the study of the dynamics within foams. A foam is a dispersion of gas droplets in a fluid. The volume fraction of the gas droplets is typically large, on the order of 0.9 or greater. As a consequence, the droplet shape is considerably deformed from spherical. Thus the droplets do not move any appreciable amount, and a foam is typically a solid, with a finite yield stress. Nevertheless, the foam still possesses internal dynamics. The size of the droplets in the foam are not constant in time; they change due to the diffusion of the gas through the fluid, out of the small droplets and into the larger ones. As this coarsening process evolves, the droplet packing conditions change. This causes stresses to be built up within the foam, which are relieved by the motion of the bubbles collapsing. These rearrangements occur randomly, but relatively rarely, throughout the sample. It is the changes in these stresses that lead to the fluctuations in the scattered light intensity that cause the decay of the DWS autocorrelation function. This can be understood by recognizing that the rearrangement involves gas bubbles, and each bubble is significantly larger than the wavelength. Therefore, each rearrangement entails the motion of the bubble interfaces over length scales that are much greater than the wavelength. Any diffusive light path that passes through one of these rearrangements will have its path length, and hence its phase, changed by a large amount, much greater than the requisite 1.4 for the DWS autocorrelation function to decay when all of the diffusive light paths have intersected at least once rearrange ment. This can be formulated more formally to account for the DWS data from foams. The key parameter that DWS measures is the rate of rearrangement in any volume unit of the foam. These rearrangements can provide new information about the dynamics of foams that lead to their unusual rheological behavior.

Conclusions

While these have been great strides in our understanding of multiple scattering light over the past several years, the applications of multiple scattering techniques to probe the structure and dynamics of disordered mesoscopic materials is still in its infancy. Much of the early work has centered on basic research studies of colloidal suspensions and other dispersions. The use of multiple scattered light has enabled new physical processes to be studied. Perhaps even more importantly, multiply scattered light is now beginning to be employed for process control applications. Finally, the most important driving force in the further development of our understanding of the use of multiply scattered light may arise from medical imaging applications. These techniques appear to be extremely promising as a noninvasive probe of the human body, particularly the brain. This may ultimately represent the true embodiment of brain waves.

References

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