Diffusive Waves in a Dilating Scattering Medium

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We consider the propagation of a diffusive wave in a scattering medium submitted to a homogeneous expansion. The light multiply scattered by a glass spheres sample is measured. We analyze the variations of the scattered light when the material and the optical wavelength are dilated. We experimentally show that an isotropic expansion of the material is equivalent to a contraction of the wavelength. Moreover, the effect of an expansion of the material on the scattered wave may be canceled by a proportional increase of the wavelength, keeping the phase of the scattered wave unchanged. Applications to the characterization of deformation of disordered materials are outlined.

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Because wave propagation through random media arises in a wide range of situations and has many potential applications, it is the focus of many experimental and theoretical studies [1,2]. Many generic behaviors are observed independently of the nature of the wave. One of these is the diffusive transport of waves [1,2], characterizing the propagation of light through a colloidal suspension, among other examples. The diffusive transport of waves is not only a unified framework to describe wave propagation or the dynamical properties of matter.

Moreover, information about the wave propagation or the variations of a scattered wave may be obtained by studying the dynamics of the material may be obtained by studying the modification of the speckle. The variations of a scattered wave following a minute deformation of disordered materials are often used to study the dynamics of matter [7]. The variations of the scattered waves under affine deformations of matter have also been discussed and used to measure deformation rates in flowing colloidal suspensions [8–11].

These two kinds of variations seem to have very different origins. However, the phase of a wave traversing the medium is a measure of the geometric distance in units of wavelength. The same variation of the phase difference between two points may be obtained either by a variation of the wavelength or by a variation of the path length. In particular, if the path length and the wavelength vary in the same proportions, the phase difference remains constant. Thus, modifying the wavelength is geometrically equivalent to expanding or contracting the medium. The purpose of this Letter is to demonstrate experimentally that this equivalence holds for diffusing waves although a large number of waves with different paths are involved in each speckle spot. In perspective of the known uses of speckle correlation in the measurement of deformations [2,12], we show that this homothetic effect permits measurements of volumetric strain and nonisotropic expansion in complex deformations of materials.

We considered experimentally the diffusive propagation of light through a disordered dielectric material made of glass beads in air. This granular material is dilated using thermal expansion. Soda lime glass beads (Potters-Ballotini) of diameter $d$ in the range 63–71 μm are placed in a spectroscopy cell (Hellma OS) of thickness $L = 5$ mm. The thermal expansion coefficients of the cell and the soda lime beads are close, making the expansion nonfrustrated. The experiments presented here were performed at temperatures $T$ between 30 and 34 °C. The sample is first slightly compacted by tapping the cell with a metallic object. The cell is then placed in a thermal enclosure controlled with a precision of a few mK. We apply five 4 °C thermal cycles before starting experiments. The sample is illuminated with a $\lambda = 633$ nm HeNe laser. After propagation through the slab, the incident beam is spread to a ~5 mm diameter spot and the transmitted scattered light is collected. The far field speckle is observed with a CCD camera. Approximately $2.5 \times 10^4$ speckle spots of size $=40$ μm are collected simultaneously. The laser wavelength $\lambda$ can be modulated without mode hop according to $(\Delta\lambda/\lambda_0) = (\Delta\lambda/\lambda_0)[1 - \cos(2\pi f_m t)]$, where $t$ is the time, $f_m = 33$ mHz the modulation frequency, and $(\Delta\lambda/\lambda_0) = 4.8 \times 10^{-7}$ the modulation amplitude, which has been measured with a Fabry-Pérot cavity. The wavelength modulation causes variations of the laser intensity which are corrected by normalizing the intensity correlation function [13]...
where $I_1$ and $I_2$ are the scattered intensities of a given pixel at two different times $t_1$ and $t_2$ and $\langle \cdot \rangle$ is an average over the recorded camera pixels.

Figure 1(b) shows measurements of the correlation function $C_f(\Delta T, \Delta \lambda/\lambda)$ between the scattered intensity $I(T, \lambda)$ at temperature $T$ and laser wavelength $\lambda$ and the scattered intensity $I(T + \Delta T, \lambda + \Delta \lambda)$ at temperature $T + \Delta T$ and wavelength $\lambda + \Delta \lambda$. The absorption of the graph is time, which corresponds to temperature as the temperature varies at a constant rate of $3.0 \times 10^{-3}$ °C/min in all experiments [thick line in Fig. 1(a)].

In a first experiment, the temperature of the material varies while the laser wavelength is kept constant. The intensity correlation function $C_f(\Delta T, \Delta \lambda/\lambda = 0)$ is the thick envelopelike line in Fig. 1(b). It is a smoothly decreasing function of the absolute temperature variation $|\Delta T|$. The correlation loss is due to the expansion of the beads, which causes a variation of the path lengths for photons traversing the sample [14]. It can be shown that it is similar to previously reported experiments where a strain field is applied to light diffusing fluids [8–11]. In a second experiment, in addition to the linear variation of temperature, the wavelength of the laser is modulated with time [fine line in Fig. 1(a)]. The intensity correlation function obtained is the oscillating curve in Fig. 1(b).

The reference image is chosen so that the wavelength modulation $\Delta \lambda$ is always positive. As long as $\Delta T$ is negative, the wavelength modulation decreases the correlation compared to the situation where $\Delta \lambda = 0$: The losses of correlation due to thermal contraction and to wavelength dilation add up. The situation is opposite when $\Delta T$ is positive, i.e., when the system is expanded: The wavelength modulation then increases the correlation. This indicates that the effect of the material expansion on the scattered wave may be compensated by an increase of the wavelength. It has been checked that, for a choice of the reference state giving a wavelength modulation which is always negative, a contraction of wavelength then compensates a contraction of the material at the same level.

The scattered electric field is $E(\lambda, T) = \sum_j E_j e^{i\Phi_j(\lambda, T)}$, with $\sum_j$ the sum over all possible paths in the material and $E_j$ the amplitude of the field for path $j$. The phase shift after propagation through the sample is

$$\Phi_j(\lambda, T) = \frac{2\pi}{\lambda} \left[ n_g(T, \lambda) L_j^g + L_j^\ell \right] + p_j \pi,$$

where $n_g(T, \lambda)$ is the refractive index of glass at temperature $T$ and wavelength $\lambda$ and $L_j^g$ (respectively, $L_j^\ell$) the length of the $j$th path through glass (respectively, air). The last term gives the integer number $p_j$ of phase shifts at the glass to air interface and is constant for a given path. The electric field correlation function is $C_E = \langle E(\lambda, T) E^*(\lambda + \Delta \lambda, T + \Delta T) \rangle$, where $\langle \cdot \rangle$ is an average over many speckle spots. The phases $\Phi_j$ are independent and, modulo $[2\pi]$, they are uniformly distributed over $[0, 2\pi]$. Consequently,

$$C_E = \sum_j |E_j|^2 e^{i\delta \Phi_j(\Delta \lambda, \Delta T)},$$

where $\delta \Phi_j(\Delta \lambda, \Delta T)$ is the phase variation with temperature and wavelength. Linear expansion of $\delta \Phi_j$ gives

$$\delta \Phi_j = \frac{2\pi n L_j}{\lambda} (\epsilon + \chi_j) = \frac{2\pi n L_j}{\lambda} \epsilon,$$

with $\chi_j = \frac{n_g - \bar{n}}{n} ([n_g - 1] \epsilon + \frac{\partial n_g}{\partial T} \Delta T + \frac{\partial n_g}{\partial \lambda} \Delta \lambda)$ and

$$\epsilon = \frac{\Delta T}{\alpha_T + \frac{\bar{n}}{n} \frac{\partial n}{\partial T}} + \frac{\Delta \lambda}{\alpha_T} \left[ -1 + \frac{\bar{n}}{n} \frac{\partial n}{\partial \lambda} \right].$$

$L_j = L_j^g + L_j^\ell$, $\eta_j = L_j^g / L_j^\ell$, is an average over all possible paths, and $\bar{n} = n_g \bar{\eta} + (1 - \bar{\eta})$ is the mean refractive index.

We considered for the derivation of (4) that the nonfrustrated expansion increases lengths inside and outside beads by the same factor $\alpha_T \Delta T$, with $\alpha_T$ the thermal expansion coefficient. Since $\epsilon$ does not depend on the path $j$, the contributions proportional to $\epsilon$ in (4) cancel simultaneously for all paths when $\epsilon = 0$. The $\chi_j$ contribution in (4) arises from the fluctuation of $n_g$ and does not cancel with $\epsilon$ in the presence of thermo-optical effects. Describing the path $j$, of length $L_j$ as $L_j / l^*$ independent segments of length $l^*$ (or $d \sim l^*$ [14]), we obtain fluctuations of $\eta_j - \bar{\eta}$ of order $\sqrt{l^* / L_j}$. In a slab geometry, $L_j \sim L_j^2 / l^*$. We then obtain that the $\chi_j$ contribution in (4) is of order $(2\pi L_j / \lambda) ([n_g - 1] \epsilon + (\partial n_g / \partial T) \Delta T + (\partial n_g / \partial \lambda) \Delta \lambda)$.

With the value of the thermo-optical effect given below and with $\Delta T = 0.1$ °C, we obtain $2\pi n L_j \alpha_T / \lambda \sim 10^{-2} \ll 2\pi$ when $\epsilon = 0$. The fluctuations $\langle \delta \Phi_j^2 \rangle \sim 10^{-4}$ are far below our experimental resolution on $C_f$. For large $\epsilon$, the $\chi_j$ contribution in (4) is small compared to $2\pi n L_j \epsilon / \lambda$.
because \( \mathcal{L}_j \gg L \). The fluctuations of \( \eta_j - \bar{\eta} \) from path to path may then be neglected in (4).

As shown in Fig. 1(b), the correlation \( C_j \) can indeed be recovered up to a value close to 1. The modulation \( \Delta \lambda / \lambda \) and temperature variations \( \Delta T \) are measured every time the correlation is recovered. Figure 2 shows the relation between these two quantities. As expected from (5), \( \Delta \lambda / \lambda \) is proportional to \( \Delta T \) when recorrelation occurs. We measured a slope \( \beta = (\Delta \lambda / \lambda) / \Delta T = 9.8 \times 10^{-6} \) K\(^{-1}\). In the absence of dispersive and thermo-optical effects, we expect \( \Delta \lambda / \lambda \) \( \Delta T \) as \( \alpha_T \). The measured value of \( \beta \) is close to the thermal expansion coefficient known for soda lime glass \( \alpha_T = 8.5 \times 10^{-6} \) K\(^{-1}\). Thermo-optical and dispersion corrections may be estimated. For soda lime of chemical composition close to the glass we used, reported values of thermo-optical coefficients are (\( \partial n_g / \partial T \) = \(-2.3 \times 10^{-6} \) [15] and \(-1.3 \times 10^{-6} \) K\(^{-1}\) [16]. We use an intermediate value \(-1.8 \times 10^{-6} \) K\(^{-1}\) in the following. The fraction \( \bar{\eta} \) of optical path length in glass is taken as 0.65 [14], close to the expected solid volume fraction. With \( n_g = 1.51 \) and \( \lambda (\partial n_g / \partial \lambda) = -0.03 \), we expect \( \beta = [\alpha_T + \bar{\eta}(\partial n_g / \partial T) / \bar{n}] / [1 - \bar{\eta}(\partial n_g / \partial \lambda) / \bar{n}] = 7.6 \times 10^{-6} \) K\(^{-1}\) in reasonable agreement with the experimental value. In a situation of homogeneous deformation, the wavelength dilation for which recorrelation is obtained is proportional to the volumetric strain sustained by the material.

The preceding analysis of experimental data concerned only the cancellation of the phase shifts \( \delta \Phi_j \) by combining a material expansion and a proportional wavelength dilation. More generally, the variation of the correlation \( C_j \) should be a function of \( \epsilon \) defined in (5). This may be checked experimentally. For this, we compute for every temperature and wavelength variations \( \epsilon = \gamma [-\beta \Delta T + (\Delta \lambda / \lambda)] \), with \( \beta \) the experimental value determined above and \( \gamma = -1 + \bar{\eta} \lambda (\partial n_g / \partial \lambda) / \bar{n} = -1.02 \). Figure 3 gathers the experimental values of the correlation function plotted as a function of the relative expansion \( \epsilon \). The points actually collapse to a master curve. This unambiguously shows that (4) and (5) describe the phase shift when wavelength and material expansions are combined. The dependence of \( C_j \) with \( \epsilon \) can be calculated using the Laplace transform \( f(p) = \int P(L) \exp[-pL] dL \) of the path length distribution \( P(L) \). Then (3) can be expressed as

\[
C_L(e) = f(i2\pi n \epsilon / \lambda).
\]  

(6)

\( f(p) \) may be calculated from the solution of the diffusion equation. For a plane-parallel slab geometry, we obtained [6]

\[
f(p) = J_0(\alpha) \frac{\sinh(z_0 \xi) + z_\sigma \xi \cosh(z_0 \xi)}{[1 + (z_\sigma \xi)^2] \sinh(L \xi) + 2 \xi z_\sigma \cosh(L \xi)}.
\]  

(7)

with \( L \) the thickness of the slab, \( \alpha \) the absorption coefficient, \( z_\sigma \) the extrapolation length, and \( z_0 \) a length of the order of the transport mean free path \( \lambda^* \). For our material. These values were obtained from measurements of light transmission through a slab [14,18,19]. We found \( \alpha^{-1} = 1.3 \) mm and \( \lambda^* = 187 \) \( \mu \)m. We use in (8) \( z_0 = \lambda^* \) and \( z_\sigma = 2 \lambda^*/3 \). The best adjustment of experimental data with (6)–(8) is found for \( (\lambda^*/6\pi n)^{1/2} = 2.0 \) \( \mu \)m in close agreement with the expected value of 2.2 \( \mu \)m.

We finally discuss the experimental observation that the correlation is not fully recovered: \( C_j \) always remains strictly smaller than unity. Experiments show that the correlation recovery depends on the preparation of the glass spheres packing. Figure 4 shows an experiment similar to the one represented in Fig. 1. The only difference is the preparation of the sample: We did not apply beforehand five temperature cycles as described above. We have plotted for comparison the correlation recovery for a more packed granular medium [i.e., the best recovery data of Fig. 1(b)]. The loss of correlation is much worse without

![Image](https://example.com/image.png)

**FIG. 2.** Variation of the wavelength vs the variation of the temperature to recover correlation. The line is a linear fit.

![Image](https://example.com/image.png)

**FIG. 3.** Intensity correlations as functions of absolute relative |\( \epsilon \)|. Dots are experimental data where thermal expansion and wavelength modulation are combined. The line is the fit with (6)–(8). Inset: Zoom with linear scale.
preparation and may be interpreted as arising from deviation from a simple isotropic expansion.

To simplify, we consider the granular material as an assembly of pointlike scatterers [20] located at positions $\mathbf{r}_p$. The refractive index is $n$, and thermo-optical and dispersive effects are neglected. When the medium expands, the motion of the scatterers is the sum of a uniform expansion $\alpha_T \Delta T \mathbf{r}_p$ and a fluctuating part $\delta \mathbf{r}_p$. The phase variation (4) along a path $j$ in the material is then $\delta \Phi_j(\Delta \lambda, \Delta T) = 2\pi n \mathcal{L}_j \epsilon / \lambda + \sum_{\mathbf{r}_p} \mathbf{q}_p \cdot \delta \mathbf{r}_p$, with $\epsilon = \alpha_T \Delta T - \Delta \lambda / \lambda$, $\sum_{\mathbf{r}_p}$ a sum over all the scatterers of the $j$th path, and $\mathbf{q}_p$ the scattering vector. When the material expansion is balanced with the wavelength expansion (i.e., $\epsilon = 0$), $\delta \Phi_j$ no longer vanishes. Nevertheless, the maximum recorrelation still leads to a measure of the volumetric strain. From the nonisotropic part, assuming that successive $\delta \mathbf{r}_p$ are independent, we obtain [12]

$$C_E(\epsilon = 0) = f \left( \frac{1}{3} \frac{k^2 \delta r^2}{r^2} \right),$$

with $k = 2\pi n / \lambda$ and $\delta r^2$ the quadratic mean of the nonisotropic deformation over the material. Inset shows the deduced values of $\delta r^2$ as a function of the temperature difference. Hence, the method leads to a measurement of the nonisotropic deformation from the maximum recovered correlation. These results are in agreement with the recently reported observation of granular compaction provoked by temperature variations [21,22]. Indeed, the presence of irreversible motion in amorphous solids [23] or granular materials [24] may be related to the departure from a locally affine displacement. Moreover, experiments in progress show that residual deformation after a temperature cycle is correlated to the amount of nonisotropic dilatation.

We demonstrated in this Letter that the principle of compensation of a geometrical expansion by an appropriate wavelength expansion persists for a diffusive wave. This principle may be used not only as a technique to determine isotropic expansions of complex materials but also to retrieve the expansion part of a more complex, including nonaffine, deformation. This principle of compensation and its use as a probe of isotropic expansion of material should remain valid for electromagnetic waves, matter waves, and sound in strongly scattering media.

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