Light transmission in porous silicon dioxide filled with liquids of different refractive indices

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Optical transmission at 532 nm from nonabsorbing disordered porous silicon dioxide has been studied experimentally. The transmission behaviors can be adjusted by filling the pores with liquids of different refractive indics, which are analyzed based on the theory of diffusion in a weak scattering regime. In our experiment, the transmission coefficient changes from a value less than 1% to one that is greater than 75%, that is, the opaque sample becomes transparent, which means that the transport mean free path of light within the material has been effectively adjusted. In addition, this method is a useful nondestructive method to derive the refractive index of an unknown bulk porous material.

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The propagation of light in multiple-scattering media has been widely discussed[1−5]. Multiple scattering can be approximated by a diffusion process, whereby the incoming light into the medium is scattered successively numerous times with an associated rapid loss of memory of the incident direction. In the diffusion regime, light is transported across the diffusive material as a random walk, with the average step being the transport mean free path (TMFP, the decay length of the average single-particle Green's function[6]) l^*, which is the main characteristic of the diffusive sample. Determination of the TMFP is a substantial fundamental problem in the physics of scattering media, which is important for an understanding of the transmission and scattering behaviors in a random system. The TMFP can be determined from measurements of the total diffuse transmission, reflection[7], and coherent backscattering[8].

In a system containing components of two refractive indices, n_1 and n_2, the light scattering depends on the shapes of the components, the spatial structures, and especially, the relative refractive index $n_\mathrm{r} = n_1/n_2$. In other words, the diffusion properties can be easily controlled by changing the concentration, the size of the scatters, and the relative refractive index. Bret *et al.* adjusted the TMFP of light by varying the concentration of spherical particles, as determined by the coherent-backscattering effect[8]. Anderson localization in gallium arsenide powders ($n_\mathrm{r} = 3.5$)[9] and a macroporous silicon two-dimensional (2D) photonic structure ($n_\mathrm{r} = 3.4$)[10] have been previously reported. Furthermore, the propagation of light in a three-dimensional (3D) random network of GaP has been studied with the ratio n_r being equal to 3.3[11].

In this letter, we experimentally studied light transport in pieces of porous silicon dioxide[12] with a volume fraction of (7.63±0.62)%. The refractive index of bulk silicon dioxide ranges from 1.42 to 1.57; therefore, the relative refractive indices of silicon dioxide to air and to liquid are smaller than 2.0, both under a weak scattering regime. According to the diffusion theory, we analyzed the relationship between transmission and thickness of our porous silicon samples. The TMFP was successfully adjusted by injecting liquid into pores, which was measured by detection of transmission. The material can be reused after evaporating the liquid. Moreover, our sample became transparent after using an appropriate index-matching liquid. Comparing the experimental results in Refs. [8,11], we obtained a greater variation of the transmission of light and the TMFP with a smaller change of the refractive index of the matching liquid. In addition, this is a convenient nondestructive method useful in deducing the refractive index of an unknown bulk porous material.

Firstly, the total transmission of 532-nm light was measured for samples with different thicknesses. The source of the incident green light was a continuous-wave diode-pumped laser. The total intensity of the transmitted diffuse light was collected with a barium sulfate integrating sphere, having a high reflectivity of 98.4% at 532 nm, and measured using a photodiode detector. Figure 1 illustrates the changes in the total transmission with the thickness, L, of porous silicon dioxide samples; the black squares in the figure are the experimental values. The absorption of silicon dioxide in the visible wavelength range is low enough to be negligible[13]. The propagation of light in optically thick, disordered scattering media can be described well by the diffusion equation in the weak scattering limit, $\mathbf{k}l_\mathrm{s} \gg 1$, where $\mathbf{k}$ is the wave vector in the medium and l_s is the scattering mean free path (SMFP, the length over which the momentum transfer becomes uncorrelated[6]). The diffusion equation neglects the interference of waves propagating along different paths, because, on average, this interference cancels out[7]. According to the diffusion theory, the total transmission in a medium is the sum of a "ballistic" contribution, $T_\mathrm{B} = \exp(-L/l_\mathrm{s})$, from photons that remain unscattered from an incident beam and a "diffuse" contribution, given by the expression

$T_D = [(1+z)-(1+z+L/l^*)\exp(-L/l_s)]/(L/l^*+2z)$[14]. Here, z is the extrapolation length ratio, which in turn depends on the diffuse reflection coefficient R, represented as $z = [2(1+R)]/[3(1-R)]$. Then, we can calculate the total transmission coefficient as follows:

$$T = T_B + T_D = \frac{(1+z)-(1-z)\exp(-L/l_s)}{(L/l^*)+2z}. \quad (1)$$

In our experiment, the raw samples are sufficiently thick to prevent the transmission of ballistic photons ($T_B \ll T_D$). Using this approximation, the exponential term in Eq. (1) becomes negligible, and T can be described by

$$T = \frac{1+z}{(L/l^*)+2z}. \quad (2)$$

The effective refractive index of porous silicon dioxide samples is between 1.029 and 1.038, when calculated from the effective-medium theory. Considering the low effective refractive index, z approximates to 0.7. For such opaque media, the transmission has no direct dependence on the scattering anisotropy. The only two relevant parameters are the sample thickness and the TMFP. Therefore, we can easily extract the value of the TMFP from the results in Fig. 1. The solid curve is the fitted result using Eq. (2), and the fitted TMFP is 10±3 μm, which is reasonable for our porous samples having average hole sizes of about 10 μm.

In the diffusive regime, the SMFP of a random medium is expected to be inversely proportional to the concentration ρ and the total scattering cross section σ_{scat} of the scatterers, represented as follows: $l_s = 1/(\rho\sigma_{\text{scat}})$[15]. According to a study on multiple scattering of light in porous materials[16], we have the following relationship between the transport cross section, σ, and the relative refractive index, n_r, of silicon dioxide to air: $\sigma \propto (n_r^2-1)^2$. In other words, the TMFP of a random medium can be modified with components of different refractive indexes in a binary system. For samples much thicker than the extrapolation length ($L \gg zl^*$), Eq. (1) shows that the relative increase of transmission matches the relative increase of l^*. As the network of the porous silicon dioxide is completely interconnected, the material is mechanically stable and can be filled with liquids of different refractive indices. Therefore, the liquid in the porous sample can be considered the background of SiO_2, which has a constant refractive index; the subsequent changes of the ratio n_r will cause the variation of l^*.

The liquids we used were mixtures of glycol and water with different weight percentages, whose refractive indices were 1.333, 1.351, 1.370, 1.390, 1.410, and 1.432. The samples with different thicknesses were fully immersed in the liquids for 10 h. Then, the infiltrated samples were taken out for measurement of transmission, that is, the interfaces of the diffusive sample were 'air-infiltrated sample–air'. The diffuse light transmission after the samples are treated is shown in Fig. 2. The black squares in Fig. 2 show the experimental results of transmission from the sample of thickness 1.95 mm. The transmission increases with the refractive index of the liquid, that is, decreases with the relative refractive index n_r. From the experimental data in Fig. 2, we can see that the maximum transmission through the infiltrated samples can reach approximately 78%, which is much larger than that of raw porous samples without any liquid. Obviously, more ballistic photons were transmitted through the treated samples, which caused the high rate of transmission of the sample. Hence, in this regime, the ballistic contribution can no longer be neglected, and the curve fitted with Eq. (1) from the diffusion theory (considering the change of z with reference to different liquids) is shown as the solid line in Fig. 2. The maximum transmission is approximately 86%, and the corresponding refractive index of the liquid is close to 1.476±0.023; in other words, the refractive index of bulk silicon dioxide is nearly 1.476. However, the maximum transmission is lower than 92%, as calculated from the Fresnel equation, which is attributed to the incomplete infiltration of our porous samples. In other words, not all the pores are occupied by the liquid and thus scattering is not totally avoided.

The same measurements have been carried out for more samples with different thicknesses, as shown in Fig. 3. From the results, we can see that, with the same index-matching liquid, the transmission increases as the sample thickness decreases. Considering the relation between

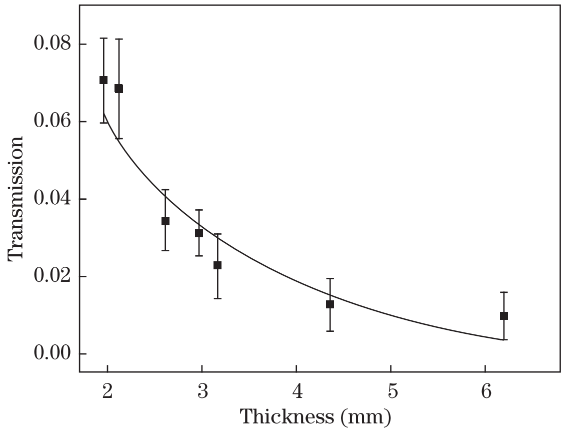

Fig. 1. Total transmission as a function of thickness of the porous silicon dioxide samples at 532 nm.

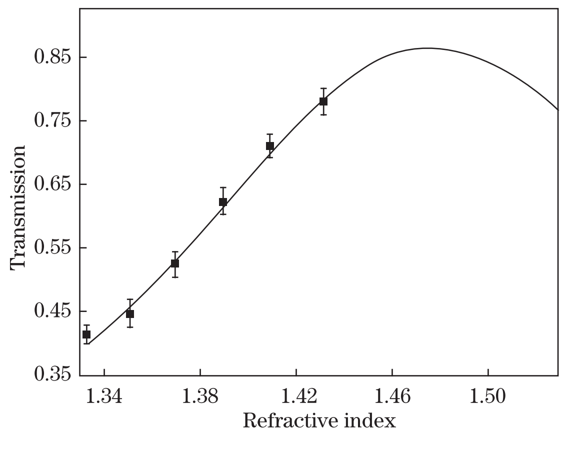

Fig. 2. Transmission of light through porous silicon dioxide samples (of thickness 1.95 mm and filled with different liquids) as a function of the refractive index of the infiltrated liquid.

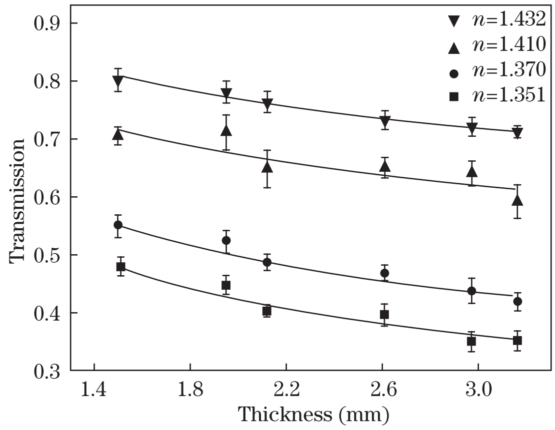

Fig. 3. Transmission of light through porous silicon dioxide filled with different mixtures as a function of sample thickness.

the cross section of transport and the relative refractive index, the refractive index of the bulk silicon dioxide in our experiments is calculated to be 1.480±0.012, based on Eq. (1), which is consistent with the result from Fig. 2.

In the above discussion, according to the diffusion theory, the ballistic contribution of transmission is assumed to undergo decay as expressed by the term $\exp(-L/l_{\mathrm{s}})$, whereas the diffuse contribution is related to the TMFP. The relationship between l_{s} and l^* is given by the expression $l^* = l_{\mathrm{s}}/(1 - \langle\cos\,\theta\rangle)$, where θ is the scattering angle. For the sake of simplicity, we consider both SMFP and TMFP to be equal, that is, $\langle\cos\,\theta\rangle = 0$. Thus, Eq. (1) can be rewritten as

$$T = \frac{(1+z) - (1-z)\exp(-L/l^*)}{(L/l^*) + 2z}. \tag{3}$$

Based on Eq. (3), we can obtain the values of l^* from the change of the total transmission in relation to the thickness of porous silicon dioxide samples infiltrated with mixtures of different refractive indices. The TMFPs of the porous samples infiltrated with index-matching liquids of different refractive indices—namely, 1.333, 1.351, 1.370, 1.390, 1.410, and 1.432—are 0.70±0.04, 0.85±0.03, 1.20±0.12, 1.81±0.18, 1.92±0.19, and 2.70±0.27 mm, respectively. The TMFP of the porous silicon dioxide slab infiltrated with pure glycol (n= 1.432) is much larger than that of the porous silicon dioxide slab without infiltration treatment.

In conclusion, we study the transmission behavior of visible light through porous silicon dioxide samples under a weak scattering regime. The transmission properties of the porous media can be controlled through infiltration with liquids of different refractive indices. With an appropriate index-matching liquid, the opaque porous medium can become transparent. In this study, the transmission coefficient changes from a value less than 1% to one that is more than 75% and the TMFP increases from 10 μm to 2.70 mm, while the the refractive indices of the liquids varies by only approximately 0.43 units. The index-matching liquid in the porous material can be subsequently removed, which makes the material reusable. Moreover, this is a convenient nondestructive method to determine the refractive index of a bulk porous material.

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