

Scattering-phase theorem: anomalous diffraction by forward-peaked scattering media

Min Xu*

Department of Physics, Fairfield University, 1073 North Benson Road, Fairfield, Connecticut 06824, USA

[*mxu@mail.fairfield.edu](mailto:mxu@mail.fairfield.edu)

Abstract: The scattering-phase theorem states that the values of scattering and reduced scattering coefficients of the bulk random media are proportional to the variance of the phase and the variance of the phase gradient, respectively, of the phase map of light passing through one thin slice of the medium. We report a new derivation of the scattering phase theorem and provide the correct form of the relation between the variance of phase gradient and the reduced scattering coefficient. We show the scattering-phase theorem is the consequence of anomalous diffraction by a thin slice of forward-peaked scattering media. A new set of scattering-phase relations with relaxed requirement on the thickness of the slice are provided. The condition for the scattering-phase theorem to be valid is discussed and illustrated with simulated data. The scattering-phase theorem is then applied to determine the scattering coefficient μ_s , the reduced scattering coefficient μ'_s , and the anisotropy factor g for polystyrene sphere and Intralipid-20% suspensions with excellent accuracy from quantitative phase imaging of respective thin slices. The spatially-resolved μ_s , μ'_s and g maps obtained via such a scattering-phase relationship may find general applications in the characterization of the optical property of homogeneous and heterogeneous random media.

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OCIS codes: (290.5825) Scattering theory; (290.5820) Scattering measurements; (180.3170) Interference microscopy; (290.7050) Turbid media.

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1. Introduction

Multiple scattering of light by random media is ubiquitous in nature. Multiple scattering by tissue, cloud, and other random media withholds direct image of such systems. Indirect characterizing and imaging these systems with multiple scattering light has attracted immense interest due to its practical importance and noninvasiveness nature. In the limit after light being scattered a sufficient number of times, light diffuses in the random medium and light diffusion is characterized by the reduced scattering coefficient μ'_s . The transport mean free path, given by the inverse of μ'_s , can be significantly larger than the distance that light travels between consecutive scattering events, μ_s^{-1} , the inverse of the scattering coefficient. Their ratio, $\mu_s'^{-1}/\mu_s^{-1}$, is

typically 10 – 100 in strongly forward-peaked scattering media such as biological tissue probed by visible or near infrared light [1]. In the other extreme, light transmitted through a thin slice of forward scattering media of thickness $L \ll \mu_s^{-1}$ suffers minimal scattering with its unscattered intensity decreasing according to the Beer's law. The phase map ϕ of the transmitted light wave can be measured using quantitative phase imaging. The two extreme cases of light propagation in random media—diffusion of multiply scattered light and transmission of minimally scattered light—has been recently suggested inherently connected first by Wang et al. [2, 3] and later by Iftikhar et al. [4]. The values of μ_s and μ'_s of the bulk media are found to be proportional to the variance of the phase and the variance of the phase gradient, respectively, of the phase map of light passing through one thin slice of the medium. This is so called “scattering-phase theorem.”

In this paper, we report first a new derivation of the scattering phase theorem and provide the correct relation between the variance of phase gradient and μ'_s . The anisotropy factor, $g \equiv 1 - \mu'_s/\mu_s$, an important parameter linked to the morphology of the scatterers in the medium, can then be derived directly from the phase map. More importantly, we show the scattering-phase theorem is the consequence of anomalous diffraction by a thin slice of forward-peaked scattering media. A set of μ_s - ϕ , μ'_s - ϕ , and g - ϕ relations are provided, for the first time, with relaxed requirement on the thickness of the slice. The condition for the scattering-phase theorem to be valid is discussed and illustrated with simulated data. The scattering-phase theorem is then applied to determine the scattering coefficient, the reduced scattering coefficient and the anisotropy factor for polystyrene sphere and Intralipid-20% suspensions with excellent accuracy from their quantitative phase maps measured by differential interference contrast microscopy. The paper ends with a discussion of the significance and applications of this scattering-phase relationship.

2. Theory

Let's consider a thin slice of random medium of thickness L illuminated by a plane wave of unit intensity. The spatially resolved phase map $\phi(\rho)$ for wave transmission is expressed as $\phi(\rho) = k \int_0^L dz m(\rho, z)$ where $k \equiv 2\pi n_0/\lambda$ is the wave number, n_0 is the background refractive index, λ is the wavelength of light in vacuum, and m is the relative refractive index at position (ρ, z) with ρ and z the lateral and axial coordinates, respectively. The fluctuation in relative refractive index $\delta m \equiv m - 1$ satisfies $\langle \delta m \rangle = 0$ where $\langle \rangle$ means the spatial average. The phase map $\phi(\rho)$ can be readily measured with quantitative phase imaging approaches [5–9].

The relation between the scattering coefficient μ_s of the bulk medium and the variance of the phase has been obtained based on the decomposition of the transmitted statistically homogeneous wave field U into its spatial average and a spatially varying component $U(\rho) = U_0(\rho) + U_1(\rho)$ and the fact that $U_0 = \langle U \rangle$ corresponds to the unscattered wave and U_1 is the scattered component [2, 10]. When the thickness of the thin slice $L \ll \mu_s^{-1}$, the intensity of the unscattered wave is expressed as $|U_0|^2 = \left| \langle e^{i\phi(\rho)} \rangle \right|^2 = \exp(-\mu_s L)$ by the Beer's law. Hence $\mu_s L = -2 \ln \left| \langle e^{i\Delta\phi(\rho)} \rangle \right|$ where $\Delta\phi \equiv \phi - \langle \phi \rangle = k \int_0^L dz \delta m(\rho, z)$. Since $|\Delta\phi| \ll 1$ as implied by $L \ll \mu_s^{-1}$, this reduces to

$$\mu_s L = \langle (\Delta\phi)^2 \rangle \quad (1)$$

if we apply the well-known cumulant expansion theorem [11] and write $\langle e^{i\Delta\phi(\rho)} \rangle = \exp(i\langle \Delta\phi \rangle - \frac{1}{2}\langle (\Delta\phi)^2 \rangle)$. The distribution of the phase needs not to follow a Gaussian distribution for Eq. (1) to be valid.

Both relations between the scattering coefficient μ_s and the variance of the phase, and the reduced scattering coefficient μ'_s and the variance of the phase gradient are the consequence of anomalous diffraction by a thin slice of forward-peaked scattering media and the requirement of

$L \ll \mu_s^{-1}$ can be relaxed. Following the treatment of anomalous diffraction by van de Hulst [12], the scattering amplitude of light into direction θ due to the thin slice is given by

$$S(\theta) = i \frac{k^2}{2\pi} \frac{1 + \cos \theta}{2} \int (1 - e^{i\Delta\phi(\rho)}) \exp(-iks \cdot \rho) d\rho \quad (2)$$

where ks is the propagation direction of the scattered light and s is a unit direction vector using the Huygens' principle [12, 13]. The presence of the thin slice alters the field on the $z = L$ plane to $e^{i\Delta\phi(\rho)}$ from 1 and hence the *scattered* wave is $e^{i\Delta\phi(\rho)} - 1$ whereas $e^{i\Delta\phi(\rho)}$ is the total wave on that plane [12]. We could replace $\cos \theta$ in Eq. (2) by 1 as scattering is forward-peaked. The scattering cross section $C_{\text{sca}} = 4\pi k^{-2} \Im S(0)$ by the optical extinction theorem is then found to be

$$C_{\text{sca}} = 2 \int (1 - \cos \Delta\phi) d\rho. \quad (3)$$

The reduced scattering cross section $C'_{\text{sca}} = k^{-2} \int (1 - \cos \theta) |S(\theta)|^2 d\Omega$ can be simplified by first writing $1 - \cos \theta = s_{\perp}^2/2$ where s_{\perp} is the projection of s on the lateral plane and rewriting C'_{sca} as

$$\begin{aligned} C'_{\text{sca}} &= \frac{1}{8\pi^2} \int ds_{\perp} \int d\rho (1 - e^{i\Delta\phi(\rho)}) \frac{d}{d\rho} \exp(-iks_{\perp} \cdot \rho) \\ &\quad \times \int d\rho' (1 - e^{-i\Delta\phi(\rho')}) \frac{d}{d\rho'} \exp(iks_{\perp} \cdot \rho'). \end{aligned} \quad (4)$$

By performing partial integration on $d/d\rho$ and $d/d\rho'$ in Eq. (4) and then integrating over s_{\perp} , we have

$$\begin{aligned} C'_{\text{sca}} &= \frac{1}{2k^2} \int d\rho \int d\rho' \left[\frac{d}{d\rho} (1 - e^{i\Delta\phi(\rho)}) \right] \left[\frac{d}{d\rho'} (1 - e^{-i\Delta\phi(\rho')}) \right] \delta(\rho - \rho') \\ &= \frac{1}{2k^2} \int \left| \frac{d}{d\rho} (1 - e^{i\Delta\phi(\rho)}) \right|^2 d\rho, \end{aligned} \quad (5)$$

which reduces to

$$C'_{\text{sca}} = \frac{1}{2k^2} \int |\nabla\phi|^2 d\rho. \quad (6)$$

As the scattering and the reduced scattering cross sections are given by $\mu_s AL$ and $\mu'_s AL$, respectively, by definition for the thin slice of area A , we obtain

$$\mu_s L = \frac{2}{A} \int (1 - \cos \Delta\phi) d\rho = 2 \langle 1 - \cos \Delta\phi \rangle \quad (7)$$

and

$$\mu'_s L = \frac{1}{2k^2 A} \int |\nabla\phi|^2 d\rho = \frac{1}{2k^2} \langle |\nabla\phi|^2 \rangle \quad (8)$$

from Eqs. (3) and (6). In addition, the anisotropy factor $g \equiv 1 - \mu'_s/\mu_s$, representing the mean cosine of the scattering angle, is given by

$$g = 1 - \frac{\langle |\nabla\phi|^2 \rangle}{4k^2 \langle 1 - \cos \Delta\phi \rangle} \simeq 1 - \frac{\langle |\nabla\phi|^2 \rangle}{2k^2 \langle (\Delta\phi)^2 \rangle}. \quad (9)$$

Equations (7), (8) and (9) constitute the main result for the scattering phase theorem. The μ_s - ϕ relation (7) reduces to the known expression (1) under the condition $\mu_s L \ll 1$, or equivalently,

$|\Delta\phi| \ll 1$. These relations share the same origin as the anomalous diffraction by optically soft particles introduced by Hulst [12] which has found wide applications in light scattering [14–18]. Equations (7), (8) and (9) are valid for forward-peaked scattering media as long as the ray does not deviate from the forward direction. The scattering-phase theorem is applicable to a slice of homogeneous or inhomogeneous medium. In the latter case, a map of μ_s , μ'_s and g can be computed from the phase map using spatial averaging over local regions rather than the whole slice.

3. Simulations and experiments

We performed simulations to validate the scattering-phase theorem for a random medium. In simulation, the fluctuation of the refractive index of the medium $R_n(\mathbf{r}) = \langle \delta m(\mathbf{r}') \delta m(\mathbf{r}' + \mathbf{r}) \rangle$ is assumed to be the Whittle-Matern correlation function [19] given by:

$$R_n(\mathbf{r}) = \langle (\delta m)^2 \rangle \gamma\left(\frac{r}{l}\right) \quad (10)$$

with

$$\gamma\left(\frac{r}{l}\right) = 2^{1-\nu} |\Gamma(\nu)|^{-1} \left(\frac{r}{l}\right)^\nu K_\nu\left(\frac{r}{l}\right) \quad (11)$$

where $K_\nu(\cdot)$ is the modified Bessel function of the second kind. The Whittle-Matern correlation function has been used extensively to model turbulence and refractive index fluctuation in biological tissue [20, 21]. The typical values are $\langle (\delta m)^2 \rangle = 0.01^2$, $l \sim 0.5 \mu m$, and $n_0 = 1.367$ for biological tissue [22, 23]. The Fourier transform of the correlation function is given by

$$\hat{R}_n(q) = \langle (\delta m)^2 \rangle \frac{\Gamma(\nu + 3/2)}{\pi^{3/2} |\Gamma(\nu)|} l^3 (1 + q^2 l^2)^{-\nu-3/2} \quad (12)$$

when $\nu > -3/2$. Light scattering by the random medium is fully described by the power spectrum of the fluctuation of the refractive index. Following [22, 24], the scattering coefficient and the reduced scattering coefficient are given by

$$\mu_s = 2\pi^{1/2} k^2 l \langle (\delta m)^2 \rangle \frac{\Gamma(\nu + 1/2)}{|\Gamma(\nu)|} \left[1 - (1 + 4X^2)^{-\nu-1/2} \right], \quad (13)$$

and

$$\begin{aligned} \mu'_s &= \pi^{1/2} l^{-1} \langle (\delta m)^2 \rangle \frac{\Gamma(\nu + 1/2)}{|\Gamma(\nu)|} \frac{1}{\nu - 1/2} \\ &\times \left[1 - (1 + 4X^2)^{-\nu-1/2} [1 + 4X^2(\nu + 1/2)] \right], \end{aligned} \quad (14)$$

respectively, in this model where $X \equiv kl$ is the size parameter.

We set the strength of refractive index fluctuation $\langle (\delta m)^2 \rangle = 0.01^2$, the correlation length $l = 0.5 \mu m$, the background refractive index of the sample $n_0 = 1.367$, and the wavelength of the incident beam $\lambda = 0.5 \mu m$ in the simulation. The random field inside a box of size $10l \times 10l \times L$ with varying thickness $L = l, 5l, 20l$, and $100l$ was simulated using RandomFields [25] with a specified spacial resolution. The phase map was generated by line integration. The gradient of the phase was computed from the phase map using the finite difference between neighboring phases. Total 15 simulations were performed for each set of parameters with their mean and standard deviation being reported hereafter.

Figure 3, from left- to right-hand direction, displays the normalized phase map $(\Delta OPL / \sqrt{\langle \delta m^2 \rangle} l)$ where the optical path length fluctuation is given by $\Delta OPL = \int_0^L dz \delta m(\rho, z)$,

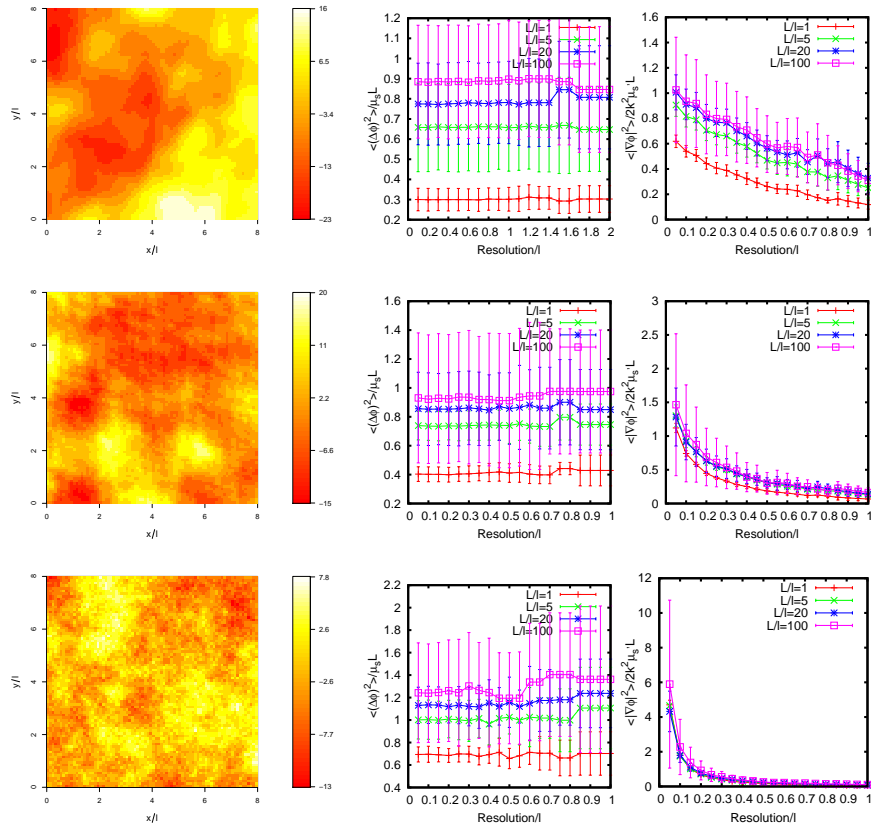


Fig. 1. The normalized phase map ($\Delta OPL / \sqrt{\langle \delta m^2 \rangle} l$), the ratio of $2 \langle 1 - \cos \Delta \phi \rangle$ over $\mu_s L$, and the ratio of $(2k^2)^{-1} \langle |\nabla \phi|^2 \rangle$ over $\mu_s^2 L$ are displayed, from left- to right-hand direction, for a thin slice of random medium of varying thickness L with the refractive index fluctuation following the Whittle-Matern correlation function of $\nu = 1.0$ (top row), $\nu = 0.5$ (middle row) and $\nu = 0.1$ (bottom row). The normalized phase map is shown for $L = 20l$.

the ratio of $2 \langle 1 - \cos \Delta \phi \rangle$ over $\mu_s L$, and the ratio of $(2k^2)^{-1} \langle |\nabla \phi|^2 \rangle$ over $\mu_s^2 L$ for various ν . The normalized phase map is shown for thin slices of thickness $L = 20l$. The scattering coefficient and the anisotropy factor for the bulk random medium are, $\mu_s l = 0.023$ and $g = 0.988$ in the case of $\nu = 1.0$, $\mu_s l = 0.015$ and $g = 0.968$ in the case of $\nu = 0.5$, and $\mu_s l = 0.0040$ and $g = 0.915$ in the case of $\nu = 0.1$, respectively. The thickness of the samples covers the range starting from $\mu_s L \ll 1$ to $\mu_s L > 1$. The two ratios $2 \langle 1 - \cos \Delta \phi \rangle / \mu_s L$ and $(2k^2)^{-1} \langle |\nabla \phi|^2 \rangle / \mu_s^2 L$ are expected to be unity according to Eqs. (7) and (8). Figure 3 shows the former ratio approaches unity when the thickness of the medium is at least $5l$. The value of μ_s can be computed from the phase map at all levels of resolution. On the other hand, the resolution matters for probing μ_s' . The latter ratio approaches unity and the best estimation for μ_s' is obtained only when the resolution is $0.1l - 0.2l$ and the thickness $L \geq 5l$. Insufficient resolution results in an underestimation of μ_s' .

We then examined the light scattering properties of polystyrene sphere and Intralipid-20% suspensions by applying the scattering-phase theorem to the quantitative phase map of respec-

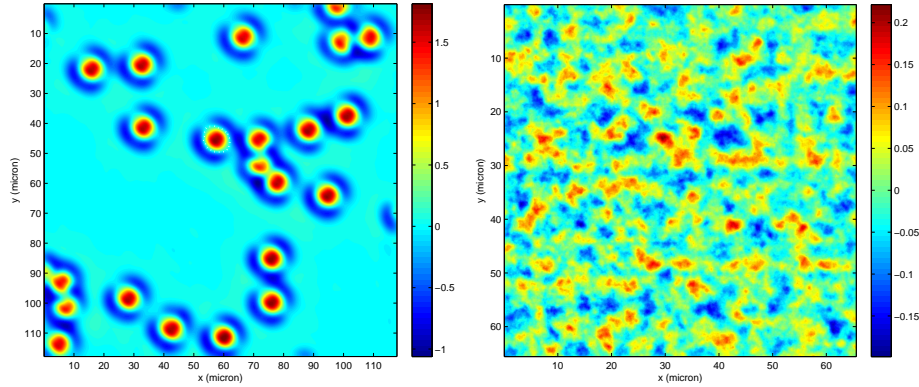


Fig. 2. The optical path length map ΔOPL for a monolayer of polystyrene sphere suspension (size: $8.31\mu m$) in water (left) and a thin film (thickness: $4\mu m$) of Intralipid-20% suspension (right).

tive thin slice measured with a differential interference contrast (DIC) microscope (Axiovert 40CFL, Zeiss). The light source was a Halogen 35W lamp filtered by a 550nm narrow-band filter. The numerical aperture for the condenser and objective (APlan 40 \times) were 0.2 and 0.5, respectively. The pixel size for the recorded images was $0.064\mu m$ using Canon 5D Mark II. The quantitative phase map for a monolayer of polystyrene sphere suspension (size: $8.31\mu m$) in water and a thin film (thickness: $4\mu m$) of Intralipid-20% suspension on a glass microscope slide were computed from in-focus and out-of-focus ($\delta z = 1\mu m$) DIC images under Köhler illumination using the transport-of-intensity approach [9]. Figure 2 shows the computed optical path length maps ΔOPL for the two samples. The scattering property for each individual spheres can be analyzed by applying the scattering-phase theorem to the region in the phase map being occupied by the sphere. For example, the region highlighted by white dash lines for the central sphere yields $\mu_s = 0.234\mu m^{-1}$, $\mu'_s = 0.0202\mu m^{-1}$ and $g = 0.91$ with an area $61.0\mu m^2$. The scattering and reduced scattering cross sections are $118\mu m^2$ and $10.2\mu m^2$. The mean scattering and reduced scattering cross sections for all the spheres contained in the displayed section are $116\mu m^2$ and $9.8\mu m^2$, respectively. These values are in excellent agreement with the theoretical prediction for a polystyrene sphere of the specified size ($C_{sca} = 125\mu m^2$, $C'_{sca} = 9.5\mu m^2$ and $g = 0.92$) computed with a Mie code [26].

The scattering and the reduced scattering coefficients for Intralipid-20% suspension are found to be $0.136\mu m^{-1}$ and $0.001\mu m^{-1}$, respectively, from the whole section displayed in Fig. 2. The former agrees with the known μ_s value ($0.139\mu m^{-1}$) whereas the latter dramatically underestimates μ'_s ($0.031\mu m^{-1}$) at 550nm [27, 28]. This behavior is expected as the characteristic correlation length for the Intralipid suspension is sub-wavelength [29] and the resolution of the phase map is insufficient to provide an accurate estimation of μ'_s directly (see Fig. 1). The quality of μ'_s estimation, however, can be significantly improved by properly taking into account light diffraction in the microscope and sharpening the phase map accordingly. This procedure yields the new value of μ'_s to be $0.022\mu m^{-1}$, agreeing reasonably well with the real value. The detail will be published elsewhere.

4. Discussion

The μ_s - ϕ and μ'_s - ϕ relations can be justified intuitively as the following. Light scattering (μ_s) depends on the fluctuation of the refractive index which emerges as the variance in the phase map for light transmission through a thin slice. Light reduced scattering (μ'_s) reflects the devi-

ation of the equal-phase wave front away from the forward direction which is described by the local tilt (gradient) in the phase for light transmission through a thin slice. Assuming the thin slice of sample of thickness L is uniformly divided into $N = L/l$ layers with l the correlation length of the random medium, $\Delta\phi$ (and $\nabla\phi$) is the summation of N independent random numbers from the N layers. Hence the spatial average $\langle(\Delta\phi)^2\rangle$ (and $\langle|\nabla\phi|^2\rangle$) scales with N rather than N^2 . These considerations lead to $\mu_s L \propto \langle(\Delta\phi)^2\rangle$ and $\mu'_s L \propto \langle|\nabla\phi|^2\rangle$. In cases such as a monolayer of scatterers of size much larger than the wavelength, the condition that $|\Delta\phi| \ll 1$ is not satisfied, the more general μ_s - ϕ relation (7) should be used whereas the μ'_s - ϕ relation remains the same provided the rays do not deviate from the forward direction (the scatterers are optically soft).

The above argument also explains that the thickness of the sample should be at least multiple l (with a sufficient large N) to obtain the values of μ_s and μ'_s correctly from the phase map as observed in the simulation. To properly compute the local tilt in the phase to obtain μ'_s with finite difference, the separation between the two points must be smaller than the size of the scattering structure. The separation at the order of $0.1l - 0.2l$ may be optimal as suggested by the simulation.

Finally, we would like to point out that the limiting form of the scattering-phase theorem when $|\Delta\phi| \ll 1$ has also been obtained previously by us using another approach [4] through analyzing the cross correlation $\langle\Delta\phi(\rho)\Delta\phi(\rho')\rangle$ between two points ρ and ρ' on the phase map for light transmission through a thin slice of a weakly scattering random medium. The scattering-phase theorem in this limit is equivalent to

$$\langle\Delta\phi(\rho)\Delta\phi(\rho')\rangle = \mu_s L - \frac{1}{2}(k\Delta\rho)^2 \mu'_s L \quad (15)$$

where $\Delta\rho \equiv |\rho - \rho'|$ is the distance between the two points [4]. Ref [2,3] presented a different expression for the g - ϕ relation. The difference originates from the scattered wave was assumed to be $e^{i\Delta\phi(\rho)}$ in our notation in Ref [2,3]. Since the presence of the thin slice alters the field on the $z = L$ plane to $e^{i\Delta\phi(\rho)}$ from 1, *the scattered wave is $[e^{i\Delta\phi(\rho)} - 1]$ whereas $e^{i\Delta\phi(\rho)}$ is the total wave* on that plane. The probability density for light scattering into direction $q = ks_{\perp}$, hence, is given by

$$P(q) = \frac{|\tilde{U}(q)|^2}{\int |\tilde{U}(q)|^2 d\mathbf{q}} \quad (16)$$

where

$$\tilde{U}(q) = \frac{1}{(2\pi)^2} \int [e^{i\Delta\phi(\rho)} - 1] \exp(-i\rho \cdot q) d\rho. \quad (17)$$

One could follow the procedure outlined in Ref [2,3] and reach the g - ϕ relation (9) if the correct probability density Eq. (16) for light scattering into direction q is used.

5. Conclusion

In summary, we have derived the scattering phase theorem and provided the correct relation between the variance of phase gradient and the reduced scattering coefficient. More importantly, the scattering-phase theorem is shown to be the consequence of anomalous diffraction by a thin slice of forward-peaked scattering media. A set of μ_s - ϕ , μ'_s - ϕ , and g - ϕ relations have been provided, for the first time, with relaxed requirement on the thickness of the slice. The condition for the scattering-phase theorem to be valid has been discussed and illustrated with simulated data. The scattering-phase theorem has been applied to determine successfully the scattering

coefficient, the reduced scattering coefficient and the anisotropy factor for polystyrene sphere and Intralipid-20% suspensions from their respective quantitative phase map of a thin slice.

The characterization of the scattering properties (μ_s , μ_s' , and g) of biological tissue and cells has been a challenging and important problem in biomedical optics [30]. This scattering-phase relationship establishes a new means to characterize the scattering properties of these samples. The spatially-resolved μ_s , μ_s' and g maps obtained via such a scattering-phase relationship will provide detailed local maps for scattering structures which may be of important diagnosis value, and may find applications in the characterization of the optical property of homogeneous and heterogeneous random media in general.

Acknowledgments

MX acknowledges Research Corporation, NIH (1R15EB009224) and DOD (W81XWH-10-1-0526) for their support.