Measuring nanoparticle size using optical surface profilers

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Abstract: Optical surface profilers are state-of-the-art instruments for measuring surface height profiles. They are not conventionally applied to nanoparticle measurements due to the presence of diffraction artifacts. Here we use a theoretical model based on wave-optics to account for diffraction-based artifacts in optical surface profilers. This then enables accurate measurement of nanoparticles size of a known geometry. The model is developed for both phase shifting interferometry and vertical scanning interferometry modes of operation. It is demonstrated that nanosphere radii as small as 12 nm, and nano-cylinder radii as small as 10-15 nm can be measured from a standard profile measurement using phase shifted interferometry interpreted using the wave-optics approach.

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References and links

1. Introduction

Nanoparticle measurement has traditionally been the domain of electron- and scanning probe microscopies, however the emergence of far-field optical nano-characterisation techniques is changing this paradigm [1–4]. Confocal microscopy, structured light and optical deconvolution have pushed the lateral resolution of far-field optical imaging systems well below the traditional Abbe diffraction limit [5–7]. Improvements in axial resolution have also been realized through techniques such as 4Pi microscopy, quantitative-phase microscopy and interferometric microscopy techniques [8–10]. With the aid of fluorescent markers, resolution approaching the single-molecule level has been demonstrated through techniques such as stimulated-emission-depletion fluorescence microscopy and photo-activated localization microscopy [1, 2, 11].

An alternative approach to optical nano-characterisation is the calculation of parameters related to size, shape, material composition or functionality of a sample object through an external measurand. A simple example is the use of interferometry to calculate nano-scale displacements by measuring optical phase. Another well-known example of this approach is dynamic light scattering (DLS); a technique where the average nanoparticle diameter in a suspension of monodisperse particles can be calculated by measuring the autocorrelation of a scattered-light signal [4]. An essential element of this approach is a physical model that accurately relates the measurand to the nanoparticle property of interest. In the case of DLS, the physical model is based on Brownian motion and classical light scattering to relate nanoparticle radius to a signal autocorrelation. These direct approaches complement imaging as they typically provide some kind of performance advantage. DLS for example is capable of measuring nanoparticle diameters down to 10 nm, well below the (marker-free) resolution limit of 90 nm achievable with direct imaging [3].

Here, we present a method to directly calculate nanoparticle size, and potentially other nanoparticle properties based on optical surface profilometry (OSP). It uses optical irradiance profiles as measurands. The feature of this method is that it retains many of the advantages conferred by far-field optical microscopy, permitting the location, visualization and characterization of single nanoparticles and nanostructures. This method is based on the application of a wave-optics model, so as to account for the diffraction-based artifacts that typically afflict optical surface profilers [12]. It will be theoretically demonstrated that this wave-optic model can permit OSP to be used for nanoparticle measurement. An experimental demonstration of this technique was recently presented and will be published in a forthcoming journal article [13].

In section 2, we present a brief summary of OSP theory for both phase-stepping interferometry (PSI) and vertical-scanning interferometry (VSI) modes, looking at both ray-optic and wave-optic approaches. In section 3, we demonstrate theoretically how the wave-optic approach can potentially be used to perform direct measurements of nanoparticle size, firstly for the simple case of an upright cylinder, and then for the case of a nanosphere. In section 4, we discuss the issues pertaining to the experimental feasibility of this technique, before presenting our conclusions in section 5.
2. Method

OSP is a well-established method of measuring 3D surface profiles over large areas. Commercial optical surface profilers have two main complementary modes of operation; phase-stepping interferometry (PSI) used for measuring flat surfaces with high accuracy [10]; and vertical-scanning interferometry (VSI) which is less accurate but can measure surfaces of arbitrary height [14, 15]. In PSI, height profiles are obtained by measuring the phase profile of a coherent light wave reflected off the object surface and equating it to a height profile. For VSI, height profiles are obtained by interfering two low-coherence (i.e. white) light waves and measuring the $z$ position where coherence in the superposition is maximum. Optical surface profilers use a standard optical microscope to image the object surface with an interferometric objective to generate the required interference fringes and a CCD camera to record irradiance profiles [16]. The best commercial optical surface profilers boast axial (‘$z$’) resolutions as low as 0.01 nm, while the lateral resolution remains limited by the Abbe diffraction limit to around 250-300 nm.

2.1 Ray-optic model

In the ray-optic model, the (scalar) field at the image plane is (for monochromatic illumination);

$$\psi(x, y) = \alpha e^{-i\phi(x, y)} + (1 - \alpha) e^{-i\phi_{ref}}, \quad (1)$$

where $\alpha$ is the fraction of the illumination field incident on the object surface, $\phi(x, y)$ is the phase profile at the object plane and $\phi_{ref}$ is the phase of the reference field. Constant terms are omitted for brevity. The first term in Eq. (1) arises from the object surface, while the second term arises from the flat reference surface and has constant phase.

2.1.1 Ray-optic model for PSI

The irradiance at the object plane is calculated by taking the modulus-squared of Eq. (1);

$$I(x, y) = \alpha^2 + (1 - \alpha)^2 + 2\alpha(1 - \alpha)\cos(\phi(x, y) - \phi_{ref}). \quad (2)$$

Letting $\phi_0 = \phi_{ref} + \delta$ and adjusting $\delta$ in steps of $\pi/2$, the phase term in Eq. (2) can be retrieved using the following algorithm

$$\phi(x, y) - \phi_0 = \tan^{-1}\left(\frac{I_{z/2}(x, y) - I_{z/2}(x, y)}{I_0(x, y) - I_z(x, y)}\right), \quad (3)$$

where subscripts denote the value of $\delta$ corresponding to each irradiance profile. Equation (3) is one of many PSI algorithms that can potentially be used [17]. Height profiles are generated by equating differences in the phase profile to path length differences,

$$z(x, y) - z_0 = \frac{1}{2k}(\phi(x, y) - \phi_0) \quad (4)$$

Due to the limited range of the inverse trigonometric functions in the PSI algorithms, only phase differences up to $2\pi$ (corresponding to a height of half of one wavelength) can be unambiguously determined.

2.1.2 Ray-optic model for VSI

Vertical-scanning interferometry (VSI) is a complementary scheme that does not suffer from the same height restrictions as PSI, at the cost of reduced speed and accuracy. VSI works by illuminating the object surface using white light and translating the object through $z$. The field at the image plane under white light illumination is
\[ \psi(x,y,\Delta z) = \int \psi_0(\omega) e^{-i\omega (\alpha e^{-i\phi(x,y,\Delta z,\omega)} + (1-\alpha) e^{-i\phi_\text{ref}(\omega)})} d\omega, \tag{5} \]

where \(\Delta z\) is the translation of the object surface and \(\psi_0\) is the amplitude of the illumination field. The corresponding irradiance is

\[ I(x,y,\Delta z) = \int |\psi_0(\omega)|^2 \left( \alpha^2 + (1-\alpha)^2 + 2\alpha(1-\alpha)\cos(\phi(x,y,\Delta z,\omega) - \phi_\text{ref}(\omega)) \right) d\omega. \tag{6} \]

Applying Eq. (4) to Eq. (6) yields

\[ I(x,y,\Delta z) = \int |\psi_0(\omega)|^2 \cos\left( \frac{2\omega}{c}(z(x,y) - \Delta z - z_\text{ref}) \right) d\omega + C. \tag{7} \]

where \(C\) is a constant. The first term of Eq. (7) is recognised as the real part of the Fourier transform of the illumination field spectrum. Evaluating this Fourier transform yields

\[ I(x,y,\Delta z) = \epsilon(\xi) \cos(a_\xi \xi) + C. \tag{8} \]

where \(\epsilon\) is the Fourier transform of \(|\psi(\omega - \omega_0)|^2\) with respect to the conjugate variable \(\xi = (2/c) (z(x,y) - \Delta z - z_\text{ref})\) and is sometimes referred to as the autocorrelation function [18]. Equation (8) consists of a constant term, a carrier wave and a modulation envelope. If \(\epsilon\) has a well-defined peak at \(\xi = \xi_0\) with a corresponding envelope peak in \(\Delta z\) at \(\Delta z_0\) then,

\[ z(x,y) - z_\text{ref} = \Delta z_0(x,y) + e^{\xi_0}/2. \tag{9} \]

That is, the position of the envelope peak depends linearly on the object surface height, \(z\). The surface profile is measured by detecting the peak in modulation as a function of position (denoted \(\Delta z_0\)) and equating it to the surface height. The constant terms in Eq. (9) are usually omitted since the absolute \(z\)-position of the object surface is arbitrary.

### 2.2 Wave-optic model

An alternative approach to ray-optics is the use of wave-optics to describe the physical model of optical surface profiler instruments. Wave-optics can account for diffraction effects and so is much more appropriate to use when applying OSP to nanoparticles. Using wave-optics, shift-invariant imaging systems can be described using linear systems theory [19]. Linear systems theory has previously been applied to optical surface profilers in the context of measuring surface roughness [20, 21], holography [22], and characterizing batwing artifacts in VSI [23]. A key feature of the wave-optic approach is that the object and image planes are no longer treated as being equivalent. Instead, the scalar field profiles at the image and object planes are related via

\[ \psi_{\text{image}}(x,y) = \psi_{\text{object}}(x,y) \ast \tau(x,y), \tag{10} \]

where \(\ast\) denotes a 2D convolution in \(x\) and \(y\), and \(\tau\) is the impulse response of the imaging system [19].

#### 2.2.1 Wave-optic model for PSI

Combining Eq. (1) and Eq. (10), the irradiance at the image plane under monochromatic illumination is

\[ I(x,y) = \left( (ae^{-i\phi(x,y)} + (1-\alpha)e^{-i\phi_\text{ref}}) \ast \tau(x,y) \right)^2. \tag{11} \]
Applying the PSI algorithm to Eq. (11) yields
\[
\phi_{\text{image}}(x, y) - \phi_0 = \text{Arg}\left(\left|e^{-i\phi(x, y)} \ast \tau(x, y)\right|\left|e^{-i\phi_0} \ast \mathcal{T}(x, y)\right|\right),
\]
(12)
where $\mathcal{T}$ denotes the complex conjugate of $\tau$ and Arg denotes the complex argument. If $\tau$ is real, then Eq. (12) becomes
\[
\phi_{\text{image}}(x, y) - \phi_0 = \tan^{-1}\left(\frac{\sin(\phi(x, y) - \phi_0) \ast \tau(x, y)}{\cos(\phi(x, y) - \phi_0) \ast \tau(x, y)}\right).
\]
(13)
Section 3 discusses how Eq. (12) and Eq. (13) can be used to determine nanoparticle size.

2.2.2 Wave-optic model for VSI

For white-light illumination (VSI mode) the field at the image plane is found by combining Eq. (6) and Eq. (10)
\[
\psi_{\text{image}}(x, y, \Delta z) = \tilde{\int}_\omega \psi_0(\omega) e^{-i\omega z} \left(\alpha e^{-\phi(x, y, \Delta z, \omega)} + (1 - \alpha) e^{-i\theta_\omega(\omega)}\right) \ast \tau(x, y, \omega) d\omega,
\]
(14)
The corresponding irradiance is
\[
I(x, y, \Delta z) = \tilde{\int}_\omega \left|\psi_0(\omega)\right|^2 \left|\left(1 - \alpha\right) \ast \tau(x, y, \omega)\right|^2 + \left|\alpha e^{-\phi(x, y, \Delta z, \omega) + i\theta_\omega(\omega)} \ast \tau(x, y, \omega)\right|^2
\]
\[+ 2\text{Re}\left[\alpha e^{-\phi(x, y, \Delta z, \omega) + i\theta_\omega(\omega)} \ast \tau(x, y, \omega)\right]\left(1 - \alpha\right) \ast \mathcal{T}(x, y, \omega)\right]\right) d\omega.
\]
(15)
where Re denotes the real part of the complex function. If $\tau$ is real then
\[
I(x, y, \Delta z) = \tilde{\int}_\omega \left|\psi_0(\omega)\right|^2 \left(\cos(\phi(x, y, \Delta z, \omega) - \phi_{\text{ref}}(\omega)) \ast \tau(x, y, \omega)\right) d\omega + C.
\]
(16)
Expanding $\tau$ as a Taylor series about $\omega_0$ gives
\[
I(x, y, \Delta z) = \tilde{\int}_\omega \left|\psi_0(\omega)\right|^2 \left(\cos\left(\phi(x, y, \Delta z, \omega) - \phi_{\text{ref}}(\omega)\right) \ast \tau(x, y, \omega)\right) d\omega
\]
\[+ \tilde{\int}_\omega \left|\psi_0(\omega)\right|^2 \left(\cos\left(\phi(x, y, \Delta z, \omega) - \phi_{\text{ref}}(\omega)\right) \ast \tau(x, y, \omega)\right) \left(\omega - \omega_0\right) \frac{\partial \tau(x, y, \omega)}{\partial \omega} \right|_{\omega=\omega_0} d\omega + C.
\]
(17)
The first term of Eq. (17) can be evaluated by bringing the convolution outside the integral and performing the same procedure as in Eq. (7) and Eq. (8) to give
\[
I(x, y, \Delta z) = \left[e(\xi) \cos(\omega_0, \xi) + C\right] \ast \tau(x, y, \omega_0)
\]
(18)
To a first-order approximation, the measured height at a given $x$ and $y$ is the peak of the modulation envelope of Eq. (18) in $\Delta z$. Higher order approximations can be made by computing higher order terms of the Taylor series in Eq. (17).

3. Results and Discussion

3.1 Characterizing upright nano-cylinders using PSI

To illustrate how the wave-optic model can be used to measure nanoparticle size, consider an upright circular nano-cylinder with a height profile
\[ z(x, y) - z_{ref} = \begin{cases} h & \sqrt{x^2 + y^2} \leq R \\ 0 & \sqrt{x^2 + y^2} > R \end{cases}. \]  

(19)

where \( R \) and \( h \) are the radius and height of the cylinder, respectively. If the effect of defocus is neglected, Eq. (4) can be used to obtain the phase profile in the object plane,

\[ \phi(x, y) - \phi_0 = \begin{cases} 2kh & \sqrt{x^2 + y^2} \leq R \\ 0 & \sqrt{x^2 + y^2} > R \end{cases}. \]  

(20)

Neglecting defocus is a reasonable approximation as the size of the nanoparticles is much less than the Rayleigh range of the objective. The phase profile measured using PSI can be found by substituting Eq. (20) into Eq. (13) and breaking up the convolution integral into regions of constant height

\[ \phi_{\text{image}}(x, y) - \phi_0 = \tan^{-1} \left( \frac{\sin(2kh) \iint_{\sqrt{x'^2+y'^2} \leq R} \tau \, dx' \, dy' + \sin(2k0) \iint_{\sqrt{x'^2+y'^2} > R} \tau \, dx' \, dy'}{\cos(2kh) \iint_{\sqrt{x'^2+y'^2} \leq R} \tau \, dx' \, dy' + \cos(2k0) \iint_{\sqrt{x'^2+y'^2} > R} \tau \, dx' \, dy'} \right), \]  

(21)

where \( \tau \) is a function of \( x-x' \) and \( y-y' \). Equation (21) shows that for an upright cylinder, the convolution integrals reduce to integrals of the impulse response over the two regions of constant height (i.e. the top of the cylinder and the underlying surface), weighted by corresponding trigonometric terms. To evaluate Eq. (21) numerically, the coherent impulse response is assumed to be that of an aberration-free imaging system with a finite pupil diameter, namely

\[ \tau_{\text{ideal}}(x, y) = J_1(r)/r, \]  

(22)

where \( J_1 \) denotes a first-order Bessel function of the first kind and \( r = NA \times k \left( x^2 + y^2 \right)^{1/2} \), where NA is the numerical aperture of the objective lens. It is conventional to normalize cylinder radius to enable more succinct presentations of results. Here, we define the normalized cylinder radius, \( R_N \) as

\[ R_N = NA \times kR. \]  

(23)

Fig. 1. Calculated \( \phi(0, 0) \) as a function of normalized cylinder radius for heights of 20 nm, 50 nm and 100 nm. The discontinuity occurs because the range of the inverse tangent in Eq. (26) is limited to returning values between \( \pm \pi \).
Figure 1 shows $\phi_{\text{image}}(0, 0)$ calculated as a function of $R_N$ using Eq. (21) for several different values of known height. $R_N$ can be determined from $\phi_{\text{image}}(0, 0)$, measured using optical surface profilometry. $R$ can then be determined using Eq. (23) without being constrained by the Abbe diffraction limit.

Measurement of $R$ is instead limited by the precision to which $\phi_{\text{image}}(0, 0)$ is known. A precision of 2.5 mrad is typical for commercial OSPs. This limits measured normalized cylinder radii down to 0.10-0.15 (depending on cylinder height) if it is assumed that reliable measurement can only be achieved if $\phi_{\text{image}}(0, 0)$ is greater than the precision of the OSP instrument. This corresponds to real cylinder radii of 10-15 nm for a NA of 0.85 and a wavelength of 514 nm. Values of $\phi_{\text{image}}(0, 0)$ that correspond to several different radii can be distinguished by making multiple measurements with objectives of different NA, or by modelling and measuring multiple coordinates in the phase profile.

3.2 Characterizing spherical nanoparticles using PSI

The approach in section 3.1 can be extended to arbitrary continuous height profiles by approximating the surface as $N$ regions, labelled $Q_j$ each with a discrete height $z_j$, where $j = 1, 2, \ldots, N$. Here, Eq. (13) becomes

$$
\phi_{\text{image}}(x, y) - \phi_b = \tan^{-1}\left(\frac{\sum_{j=1}^{N} \sin\left(2k(z_j - z_{\text{ref}})\right) \iiint_{Q_j} \tau(x'-x, y'-y) \, dx' \, dy'}{\sum_{j=1}^{N} \cos\left(2k(z_j - z_{\text{ref}})\right) \iiint_{Q_j} \tau(x'-x, y'-y) \, dx' \, dy'}\right),
$$

(24)

This approximation becomes exact when $N$ approaches infinity, in which case the summation terms in Eq. (24) become Lebesgue integrals;

$$
\phi_{\text{image}}(x, y) - \phi_b = \tan^{-1}\left(\int_{z_{\min}}^{z_{\max}} \sin\left(2k(z_j - z_{\text{ref}})\right) \int_{Q(z)} \tau(x'-x, y'-y) \, dx' \, dy' \, d\mu\right),
$$

(25)

where the measure, $\mu$ is given by

$$
\mu(x, y, z) = \iiint_{Q(z)} \tau(x'-x, y'-y) \, dx' \, dy',
$$

(26)

where the integration region, $Q$, varies as a function of $z$. For a spherical nanoparticle the height profile can be parameterized as

$$
z(x, y) = z_{\text{ref}} + \left\{\begin{array}{ll}
\sqrt{x^2 + y^2} & x^2 + y^2 \leq \rho \\
0 & x^2 + y^2 > \rho
\end{array}\right.
$$

(27)

where $\rho$ is the radius of the sphere. The normalized sphere radius is defined as

$$
R_N = NA \times k \rho.
$$

(28)

Figure 2 shows $\phi_{\text{image}}(0, 0)$ calculated as a function of $\rho_N$ using Eq. (24) for $N = 1, 5, 20, 50$ and 200. Values of $\rho_N$ down to 0.12 can be measured, corresponding to $\rho$ as small as 12 nm for a NA of 0.85 and a wavelength of 514 nm. The function shown in Fig. 2 exhibits three distinct regions; (i) a monotonically increasing region up until $\rho_N = 0.6$, (ii) a dip region between $\rho_N = 0.6$ and $\rho_N = 1.2$ where $\phi_{\text{image}}(0, 0)$ decreases with $\rho_N$ (even becoming negative), and (iii) a region from $\rho_N = 1.2$ onward which is monotonically increasing. As $\rho_N$ becomes large $\phi_{\text{image}}(0, 0)$ converges to a value of $4k\rho$, the expected value if diffraction effects are ignored. This convergence is quite slow; for a NA of 0.85 and $\rho_N = 5.2$, $\phi(0, 0)$ is only 70% of...
The dip region arises because of sign changes in the numerator and denominator of Eq. (24). The existence of this dip region can manifest as PSI artifacts in optical surface profiler systems where elevations appear as depressions and vice-versa.

3.3 Characterizing upright nano-cylinders using VSI

VSI can also be applied to measuring nano-particle size, though the analysis is somewhat more complex owing to the white-light illumination. In order to better illustrate the basic features of VSI when using this nanoparticle measurement technique, a series of approximations are made. Firstly the modulation envelope is taken to be that described by Eq. (18), i.e. only the first term of the Taylor series is included. The impulse response function is taken to be

\[ \tau_{\text{ideal}}(x, y) = J_1(r)/r \]

as before, except here \( r = NA \times k_0 \left( x^2 + y^2 \right)^{1/2} \), where \( k_0 \) is the central wavenumber of the illumination field spectrum. The modulation envelope (denoted \( I_{\text{env}} \)) is approximated from Eq. (18) to be

\[ I_{\text{mod}}(x, y, \Delta z) = \mathcal{E}(\xi) * \tau(x, y, \omega_0). \] (29)

Finally, we take the irradiance spectrum of the white-light illumination to be a Gaussian function with central frequency \( \omega_0 \) and 1/e² width \( \Delta \omega \). The Fourier transform is then

\[ \mathcal{E}(\xi) = e^{-4(4\Delta \omega)^2 ((x, y) - (x_0, y_0))/2\xi^2}. \] (30)

Taking the height profile of an upright nano-cylinder (Eq. (19)) and applying it to Eq. (29) yields

\[
I_{\text{mod}}(x, y, \Delta z) = \mathcal{E}(2(h-\Delta z)/c) \int_{\sqrt{x^2+y^2} \leq R} \tau(x'-x, y'-y) dx' dy' \\
+ \mathcal{E}(2(0-\Delta z)/c) \int_{\sqrt{x^2+y^2} > R} \tau(x'-x, y'-y) dx' dy' \] (31)

In the ray-optic model, there is only one modulation envelope for each point in \( x \) and \( y \). In the wave-optic model by contrast, each point in \( x \) and \( y \) will consist of multiple modulation envelopes; one for each height present in the height profile, with an amplitude that depends on the integral of \( \tau \) over that corresponding height region. Since there are multiple modulation

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Fig. 2. Calculated \( \phi(0, 0) \) as a function of \( \rho_N \) for an approximated height profile with \( N = 1, 5, 20, 50 \) and 200. The curve corresponding to \( N = 50 \) and \( N = 200 \) almost completely match, indicating good convergence is achieved for around 50 partitions. The inset shows the same curve over a smaller range for \( \rho_N \) up to 0.3.
envelopes, the total envelope possesses multiple peaks, in which case identifying the peak can become ambiguous.

![Graph showing calculated I_{mod} vs \Delta z for different cylinder heights.](image)

**Fig. 3.** Calculated I_{mod} vs \Delta z for different cylinder heights for \Delta \omega = 1.29 \times 10^{15} and \omega_0 = 3.67 \times 10^{15}.

Figure 3 shows how the envelope evolves as the height is increased, starting as a single merged envelope when \( h \) is small, before splitting off into two envelopes, one due to light reflecting from the top of the cylinder and the other due to light reflecting from the underlying surface. Altering the cylinder radius changes the relative magnitude of each envelope, which alters how the position of the peak varies with height, as shown in Fig. 4. Two distinct regimes are evident in Fig. 4. The first regime occurs for larger \( R_N \) (Eq. (23) where the second envelope (due to light reflected from the top of the cylinder) dominates the first, resulting in a continuous transition as the peak splits. The second regime occurs for smaller \( R_N \) where the first envelope (due to light reflecting from the underlying surface) dominates, resulting in a discontinuous transition as the peak splits. For the curve corresponding to the first regime (\( R_N = 1.56 \)) the position of the highest envelope is monotonically increasing, while for the curve corresponding to the second regime (\( R_N = 1.30 \)) it is not.

![Graph showing calculated \( \Delta z \) as a function of cylinder height.](image)

**Fig. 4.** Calculated \( \Delta z \) as a function of cylinder height. Solid and dashed lines indicate the position of the first and second modulation envelope peaks respectively. The dotted curve indicates the ideal measurement where the peak position is equal to the cylinder height.
3.4 Characterizing spherical nanoparticles using VSI

As with PSI, the VSI case can be generalized to arbitrary continuous height profiles by approximating the surface as \(N\) regions, labelled \(Q_j\), of height \(z_j\) where \(j = 1, 2, \ldots, N\). Equation (31) can then be generalized as

\[
I_{\text{mod}}(x, y, \Delta z) = \sum_{j=1}^{N} \varepsilon_j \left( 2(z_j - \Delta z)/c \right) \int_{Q_j} \tau(x' - x, y' - y) \, dx' \, dy'.
\]  

(32)

For the limit where \(N\) approaches infinity (i.e. for a continuous height profile), Eq. (32) becomes

\[
I_{\text{mod}}(x, y, \Delta z) = \int_{\xi_{\min}}^{\xi_{\max}} \varepsilon(\xi) \, d\mu,
\]  

(33)

where \(\xi_{\max}\) and \(\xi_{\min}\) are the values of \(\xi\) corresponding to the maximum and minimum in \(z(x, y)\) respectively. Figure 5 shows the modulation envelopes calculated for a spherical nanoparticle with a height profile given by Eq. (27) at the origin.

![Fig. 5](image)

In the specific case of the nanosphere, it is helpful to define a normalized \(\Delta z\)-coordinate as

\[
\Delta z_N = NA \times k \Delta z.
\]  

(34)

The normalized peak position is defined as the peak position in these normalized coordinates. Figure 6 shows how \(\Delta z_N\) varies with \(\rho_N\). Two distinct regions are evident. The first region, bounded by \(0 \leq \rho_N \leq 1.64\) exhibits a single peak comprised of modulation envelopes from both surface and nanosphere regions and so is termed a hybrid peak. In this region the peak position of the modulation envelope exhibits a slow, monotonic increase with increasing \(\rho_N\). Here, the difference between the calculated value and the true normalized height \((2\rho_N)\) is substantial, with a difference exceeding two orders of magnitude for some values of \(\rho_N\). In the second region where \(\rho_N > 1.64\), the modulation envelope becomes dominated by modulations envelopes from the nanosphere regions and so is termed the sphere peak. In this region the measured peak position is much closer to the true sphere height, however there is a constant residual error which exists because the modulation envelope from the topological centre of...
the nanosphere cannot be truly isolated; modulation envelopes from other nanosphere regions will always place a downward bias on the peak position of the total modulation envelope. For \( \rho_N > 2.4 \) there exists a distinct peak due to the modulation envelope from the surface region and is included only for completeness.

![Graph](image)

Fig. 6. Calculated normalized peak position of the modulation envelope as a function of \( \rho_N \).

Sphere and surface peaks are peaks dominated by sphere and surface terms respectively, while hybrid peaks are those where sphere and surface terms have significant contributions.

The precision of optical surface profilers is typically around 0.3 nm when operating in VSI mode. Assuming that a \( \Delta z \) greater than 0.3 nm is required for reliable measurement, \( \rho_N \) as small as 0.19 can potentially be measured using VSI. This corresponds to \( \rho \) as small as 19 nm for an NA of 0.85 and a central illumination wavelength of 514 nm.

4. Discussion

Given that there are a number of conditions and approximations for implementing this nanoparticle measurement technique, it is worth devoting discussion to the issue of practical feasibility.

The most stringent requirement of this technique is that of an idealized surface; as real surfaces will possess some degree of surface roughness. The question is how sensitive \( \phi_{image} \) precision is to real surface roughness. Close inspection of Eq. (21) and Eq. (24) reveal \( \phi_{image} \) to essentially depend on a weighted average. The effect of random height variations are thus expected to cancel on average and so degradation in measurement precision of \( \phi_{image} \) is expected to be minor for typical amounts of surface roughness (and indeed, the method has been demonstrated experimentally - see footnote on p.2). We are currently in the process of developing ways to quantify the sensitivity of \( \phi_{image} \) precision to surface roughness.

Assumption of an idealized surface also introduces constraints on nanoparticle density. Nanoparticles must be sufficiently dispersed so that the signal from one nanoparticle does not affect the measurement of its neighbors. There is potential to mitigate this constraint by modelling the surface as double nanoparticles, or other, larger nanoparticle conglomerates, although this increases the number of parameters required to parameterize the surface.

Another practical consideration arises is when the surface is composed of more than one material, such as a metallic nanoparticle on silica glass. In this case the phase and amplitude of the impulse response must be modified in accordance with Fresnel reflection equations.

A final note: many of the approximations used herein are not fundamental to the proposed technique, rather they exist to simplify the analysis. The assumed form of the object field in Eq. (4) for example could be replaced by a form calculated directly from Maxwell's equations without altering the general analytical procedure. As with any other measurement technique, accuracy and precision is dependent on the quality of the approximations used.
5. Conclusion

In this paper, we have demonstrated that OSP can be extended to measurement of nanoparticle size, enabled through a wave-optic model relating measured phase to nanoparticle characteristics. Such a model has been presented for both the PSI and VSI modes of operation. It is shown that nanospheres with radii as small as 12 nm can be measured with PSI, and as small as 19 nm can be measured for VSI, using a standard OSP instrument combined with this new analytical approach. Future work will be focused on extending this technique to other nanoparticle shapes and geometries (such as double-particles), quantifying the sensitivity of phase measurements to surface roughness and developing a more general vector-based approach with object fields computed directly from Maxwell's equations.

Acknowledgment

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