Accurate determination of microparticle size using Fourier transform of light scattering spectrum over wavenumber

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We discovered that the far-field light scattering spectra of microparticles over wavenumber at a certain angle could be decomposed into periodic components, with an oscillation frequency linearly dependent on the particle size at each angle. Based on this observation, we propose and experimentally demonstrate a new Fourier transform technique for microparticle size determination. This technique is simple, fast, robust in its data processing algorithm, and flexible in its detection system. © 2007 Optical Society of America

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Elastic light scattering has been used for efficient and accurate measurement of the geometrical properties (size, distribution, concentration, etc.) of microparticles, which is essential for many biomedical applications, including early cancer detection,[1] cellular morphology exploration,[2] and bacteria size determination.[3] Analyzing the angular scattering patterns[2,4] and spectral scattering patterns[5,6] are two primary techniques for microparticle characterization using light scattering techniques.

At present, most of those methods based on light scattering spectroscopy are being investigated in a backscattering geometry, since it has been shown that the spectrum over wavenumber ($1/\lambda$, with λ being the wavelength) of backscattered light has a periodic component with an oscillation frequency proportional to the particle size.[5] However, certain data fitting algorithms based on lookup tables are still required, which can be quite time consuming.[6]

In this Letter we report observation of periodicity that exists in the scattering spectrum at any angle and, additionally, that the spectrum can be decomposed into many components with different oscillation frequencies, each with linear dependence on the particle size. For most angles there is one oscillatory component predominant over the others, which we call the "major" oscillatory component. The Fourier transform is used to separate these oscillatory components. By isolating the regime of the major oscillation in the Fourier domain and inversely mapping the position of the peak in the Fourier domain (corresponding to the oscillation frequency) to the particle size, we developed a simple and fast particle sizing technique with a robust data processing algorithm and a flexible detection system.

The scattering spectrum of a spherical particle can be calculated analytically by using Mie theory.[7] Consider a dielectric sphere of diameter d and refractive index n_1 in a surrounding medium of refractive index n_0. For a nonpolarized plane wave of intensity I_{in} and wavelength λ (defined in vacuum) incident on this particle, the far-field scattering intensity I_{sca} at scattering angle θ can be expressed[8] as

$$I_{\text{sca}}(\theta,\lambda,d,n_0,n_1) = \frac{\lambda^2}{(n_0\pi)^2 r^2}(|S_1(\theta,x,m)|^2 + |S_2(\theta,x,m)|^2)I_{\text{in}}. \quad (1)$$

Here r is the distance from the scatterer to the detector, and S_1 and S_2, obtained via Mie calculations, are scattering amplitudes for polarizations perpendicular and parallel to the scattering plane, respectively. They are dependent on the scattering angle θ, a dimensionless size parameter $x=n_0\pi d/\lambda$, and the refractive index ratio $m=n_1/n_0$.

In this Letter we investigate the scattering spectra of polystyrene suspensions, i.e., dielectric microspheres ($n_1=1.59$) in water ($n_0=1.33$). Figure 1(a) shows the calculated spectra over wavenumber $1/\lambda$ for suspensions of 10 μm (diameter) particles at scattering angles of 10°, 30°, and 60°, with unpolarized incident light. In each spectrum there exists an easily observable ripple. It has been shown that the oscillation of this ripple, which is generated by the combined effects of diffraction and resonance in the dielectric spheres, is periodic.[8] Furthermore, aside from the prominent major ripple, there are more oscillatory components: a slow background oscillation and higher-order ripples with fast oscillations. Note that in Eq. (1) the particle diameter d is tied to wavenumber $1/\lambda$ by multiplication into a dimensionless size parameter, $x=n_0\pi d/\lambda$. This implies that the oscillation frequencies are proportional to the particle size. Figure 1(b) illustrates such linear relationships. The Fourier transforms of downshifted spectra (to annihilate the dc offset) versus $1/\lambda$ ($0.001-0.0025\ \text{nm}^{-1}$) are plotted for particle sizes in a range from 5 to 20 μm. The major ripple corresponds to the bright straight line, while the background oscillation and higher-order ripples generate other observable straight lines.

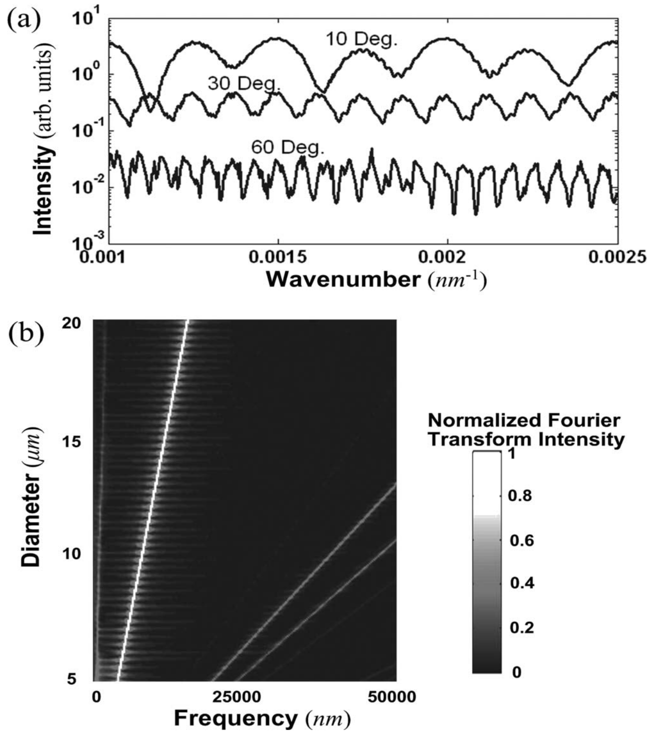

Fig. 1. (a) Calculated scattering spectra as a function of wavenumber 1/λ at different scattering angles for 10 μm polystyrene particles. (b) Linear evolution of Fourier transform of spectrum over 1λ with increasing particle size, scattering angle θ fixed to 30°. Note that the “frequency” here is the Fourier transform variable in Eq. (2).

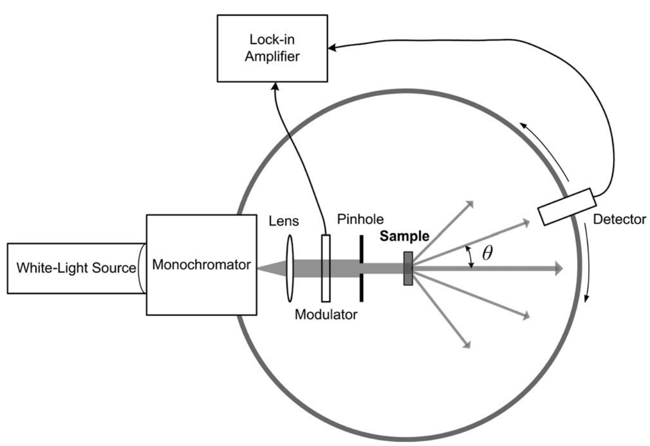

Fig. 2. Schematic of the angle and wavelength scanning detection system.

We have developed a new particle sizing technique using the Fourier transform based on this phenomenon. The major oscillatory component in a scattering spectrum is well isolated from other components in the Fourier domain, and its corresponding peak position can be linearly mapped to particle size. As the spectrum measurements are usually taken over wavelength, we need to convert the spectrum over wavelength λ into the spectrum over 1/λ before applying the Fourier transform. Consider a wavelength sampling sequence $\lambda_1, \lambda_2, \ldots, \lambda_n$ and a spectrum $I(\lambda)$. Then the Fourier transform of the corresponding spectrum over 1/λ is

$$\tilde{I}(f) = \sum_{j=1}^{n-1} \frac{1}{2}\left\{[I(\lambda_j) - C]\exp\left(-i\frac{2\pi}{\lambda_j}f\right) + [I(\lambda_{j+1}) - C]\exp\left(-i\frac{2\pi}{\lambda_{j+1}}f\right)\right\}\left(\frac{1}{\lambda_j} - \frac{1}{\lambda_{j+1}}\right), \quad (2)$$

where C is introduced to annihilate the dc offset of the spectrum and is given by

$$C = \left[\frac{1}{2}\left(\frac{1}{\lambda_1} - \frac{1}{\lambda_n}\right)^{-1}\right] \sum_{j=1}^{n-1} [I(\lambda_j) + I(\lambda_{j+1})]\left(\frac{1}{\lambda_j} - \frac{1}{\lambda_{j+1}}\right). \quad (3)$$

The experimental setup we used to measure the light scattering spectra is shown in Fig. 2. The white-light source is a tungsten lamp, incorporating a monochromator and a lens to provide collimated wavelength-tunable incident light. A detector is

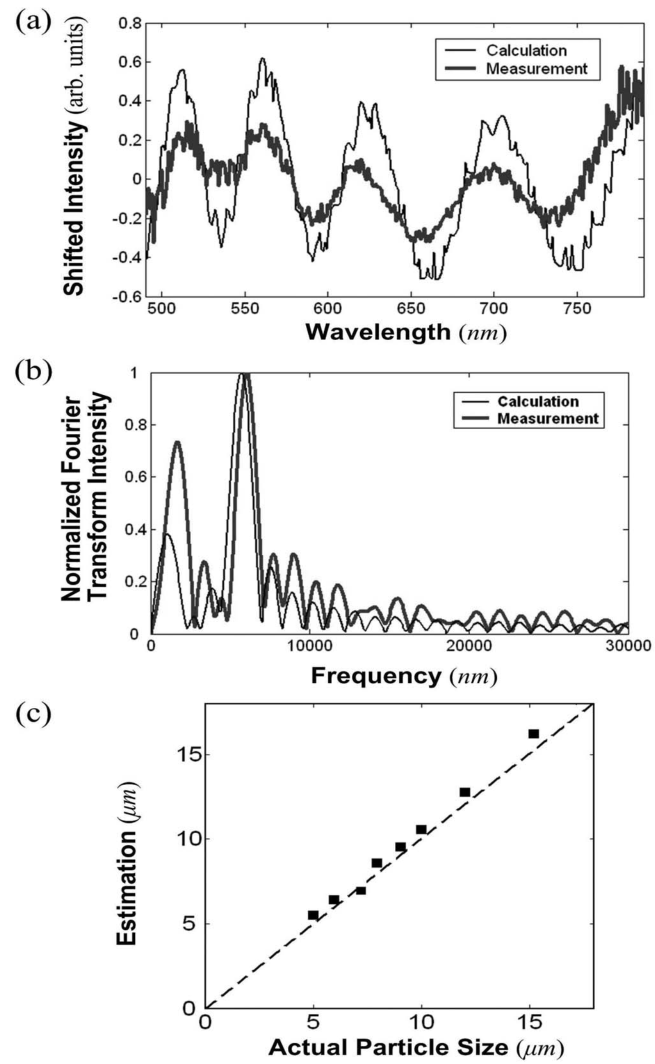

Fig. 3. (a) Calculated and measured scattering spectra for the suspension of 10 μm particles, detection angle fixed at 25°. (b) Fourier analyses for the curves in (a). Note that the “frequency” here is the Fourier transform variable in Eq. (2). (c) Particle size determination for a set of suspensions.

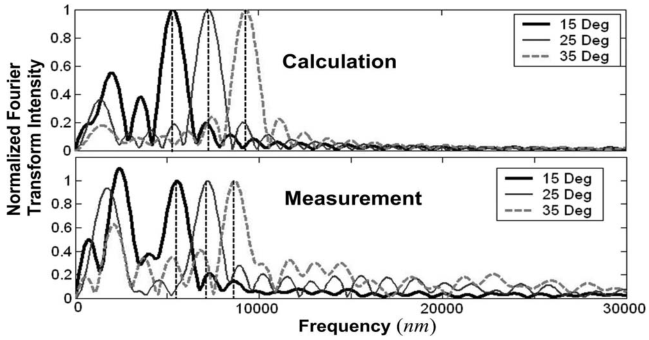

Fig. 4. Fourier analyses of calculated and measured scattering spectra for 12 μm particles at detection angles of 15°, 25°, and 35°.

mounted directly onto the arm of a rotating motor. A modulator (light chopper) and a lock-in amplifier are used to improve the signal-to-noise ratio of the detection system. The suspension samples are contained in thin quartz cuvettes of 2 mm thickness.

To verify the performance of this technique, we measured the scattering spectra (wavelength scanned from 490 to 790 nm in 1 nm intervals) of a number of polystyrene suspensions of different particle sizes, which were dilute enough to validate the single-scattering model. Due to the refraction at the water–air interface, the detection angle θ determines the scattering angle θ' by Snell's law ($\sin\theta' = \sin\theta/n_0$). Figure 3(a) shows the theoretical and experimental downshifted spectra for the suspension of 10 μm particles, with corresponding Fourier transforms shown in Fig. 3(b). Note that the major peak derived from measured data is not as dominant as from theoretical calculations, while higher-order peaks are not observable at all. The explanation depends on several factors: the probe collects light from a small range of scattering angles because of the nonzero spot area of scatterers and the numerical aperture of the detector, there is a small nonuniformity of particle sizes in each sample, and multiple scattering cannot be neglected completely. All these factors combine to smooth away the higher-order ripples and decrease the oscillation amplitude of the major ripple while having little effect on the slow background oscillation. However, since the oscillation frequencies of these components remain consistent, the precision of the particle size estimation is minimally affected, making this technique robust. The particle size estimations for a set of suspensions, based on the linear relation between the major frequency peak and the particle size, are shown in Fig. 3(c), where the average percentage error is less than 5%.

The oscillation frequencies are also angle dependent. For forward-scattered light (scattering angle less than 90°) investigated in this work, the spectrum at larger angles typically has faster oscillations but lower intensity. This is demonstrated in Fig. 4, which shows the theoretical and experimental spectra at different scattering angles. Note that the increasing peak frequencies at higher scattering angles allow for better discrimination of particle sizes, but the reduction in strength of these peaks requires more sensitive detection.

Compared with other particle size characterization techniques, our technique uses a simple setup and a fast data processing algorithm. Furthermore, as found in experiments, the discrepancy between corresponding measured and calculated spectra usually results in the variation of relative amplitude of each oscillatory component, while oscillation frequencies are not affected. This makes our technique robust in that the precision of estimation is well preserved even under less-than-ideal experimental conditions. In addition, the detection angle in our system is flexible, which enables us to employ the angular dependence of the oscillation frequencies to obtain optimum discrimination of particle size based on the available detection capability. The main limitation of this technique is the sensitive dependence of the peak position to the detection angle, which makes it important to obtain accurate angular information. However, this effect can be minimized by initial calibration of the system with precisely known particle sizes.

In conclusion, we present here a Fourier transform method of microparticle size determination using the oscillatory features of scattering spectra, which avoids the complex data fitting algorithms and sophisticated setups that are conventionally used. We show that the linear dependence of the particle size on the major peak in the Fourier transform of the scattering spectrum can be used to estimate particle sizes with good accuracy. This simple, robust, and flexible technique is competitive in many ways and is potentially applicable to several biomedical areas, especially those related to cellular morphology.

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References

1. V. Backman, M. B. Wallace, L. T. Perelman, J. T. Arendt, R. Gurjar, M. G. Muller, Q. Zhang, G. Zonios, E. Kline, J. A. McGilligan, S. Shapshay, T. Valdez, K. Badizadegan, J. M. Crawford, M. Fitzmaurice, S. Kabani, H. S. Levin, M. Seiler, R. R. Dasari, I. Itzkan, J. Van Dam, and M. S. Feld, Nature **406**, 35 (2000).
2. J. R. Mourant, T. M. Johnson, S. Carpenter, A. Guerra, T. Aida, and J. P. Freyer, J. Biomed. Opt. **7**, 378 (2002).
3. A. Katz, A. Alimova, M. Xu, E. Rudolph, M. Shah, H. E. Savage, R. Rosen, S. A. McCormick, and R. R. Alfano, IEEE J. Sel. Top. Quantum Electron. **9**, 277 (2003).
4. A. Wax, C. H. Yang, R. R. Dasari, and M. S. Feld, Opt. Lett. **26**, 322 (2001).
5. L. T. Perelman, V. Backman, M. Wallace, G. Zonios, R. Manoharan, A. Nusrat, S. Shields, M. Seiler, C. Lima, T. Hamano, I. Itzkan, J. Van Dam, J. M. Crawford, and M. S. Feld, Phys. Rev. Lett. **80**, 627 (1998).
6. V. Backman, G. Gurjar, K. Badizadegan, I. Itzkan, R. R. Dasari, L. T. Perelman, and M. S. Feld, IEEE J. Sel. Top. Quantum Electron. **5**, 1019 (1999).
7. G. Mie, Ann. Phys. **25**, 377–445 (1908).
8. W. T. Grandy, *Scattering of Waves from Large Spheres* (Cambridge, 2000).