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THE CAUSE OF CHARACTERISTIC LENGTHS IN SCATTERING CURVES

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ABSTRACT. This work explains the cause of crossovers in the power-law structure of scattering curves for spherical and nonspherical particles. For spheres, the crossovers have been empirically correlated with length scales of the particle, e.g., radius. Here, a technique called phasor analysis will show that destructive interference within the particle causes the crossovers. Nonspherical particles will also be investigated and found to display similar behavior, and several practical implications of the crossovers will be presented.

1. Introduction

The scattered intensity I in the far-field zone of a spherical particle is known to display an overall power-law structure [1]. This structure is seen when I is expressed in terms of the scattering wave vector $q = 2k\sin(\theta/2)$, rather than the polar angle θ. Patterns appear in the evolution of this structure as the particle radius R and refractive index m vary. More generally, the evolution is governed by the phase shift parameter $\rho = 2kR\mathrm{Re}\{m-1\}$, rather than R and m independently, i.e.,

$$I(\theta) \simeq \begin{cases} (qR)^0 & 0 \leq qR \lesssim \pi/2, \\ (qR)^{-2} & \pi/2 \lesssim qR \lesssim \rho \text{ if } \rho > 1 \\ (qR)^{-4} & \rho \lesssim qR. \end{cases} \tag{1}$$

An example of these patterns is shown in Fig. 1. The transition between power-laws is called a crossover, two of which are displayed by spherical particles; the Guinier crossover at $qR \simeq \pi/2$ and the ρ crossover at $qR \simeq \rho$. Because q has units on 1/length, Sorensen *et al.* propose that features in $I(qR)$ identifying single points for q represent length scales of the particle [1]. Note that the patterns relate only to the overall behavior of the curves and do not include backscattering enhancements such as glory.

Equation (1) and the crossovers have been established empirically: Mie theory is used to generate I-curves for a variety of particles, which are then collectively studied to identify commonalities using the unitless quantities kR, qR, and ρ. Because of this empirical basis, it is not clear *why* the patterns occur or what their physical significance and possible practical utility may be. The work presented here will explain the patterns' cause and

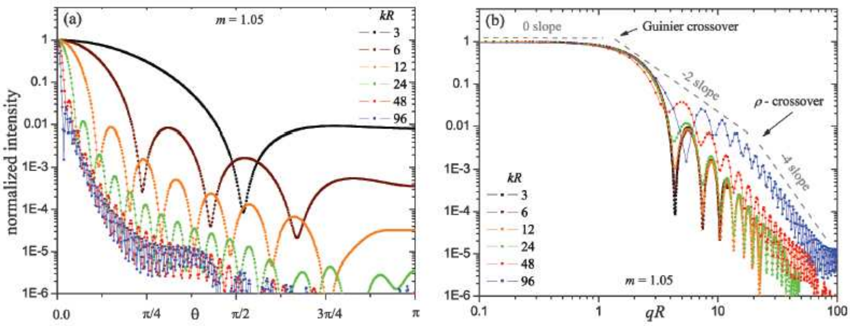

Figure 1. Power-law patterns for spherical particles. Plot (a) shows $I(\theta)$ for particles with $m = 1.05 + 0i$ and multiple size parameters kR. Plot (b) shows the *same* curves plotted as a function of qR along with the power-law envelopes and crossovers.

reveal that the crossovers are related to characteristic lengths associated with the particle interior. Although not presented in this abstract due to space limitations, the explanation will be extended to nonspherical particles using the DDA where similar conclusions are found. The interested reader can also see [2].

2. Phasor analysis

Consider a spherical particle illuminated by a linearly polarized plane wave. Given that $\mathbf{r}$ is confined to the far-field zone on a circular contour of radius R_l in the horizontal scattering plane, $\mathbf{E}^{\text{sca}}$ can be expressed as the superposition of spherical waves, or wavelets for short, [3]

$$\mathbf{E}^{sca}(\theta) = \frac{\exp(ikR_l)}{R_l} \lim_{\Delta V \to 0} \sum_i z_i(\theta)\, \hat{\mathbf{x}}, \tag{2}$$

where

$$z_i(\theta) = \frac{k^2}{4\pi}(m^2 - 1)E_x^{\text{int}}(\mathbf{r}_i) \exp(-ik\hat{\mathbf{r}} \cdot \mathbf{r}_i)\, \Delta V. \tag{3}$$

In essence, this is just Huygens' principle, except generalized to full vector form and taking into account the particle's internal field $\mathbf{E}^{\text{int}}$. Equation (2) shows that each volume element ΔV contributes a wavelet to $\mathbf{r}$, the (complex-valued) amplitude of which is given by the phasor z_i, in Eq. (3). The scattered intensity I can then be obtained directly from Eq. (2).

The advantage of expressing $\mathbf{E}^{\text{sca}}$ in terms of phasors is that they enable a *graphical* way to understand how the wavelet superposition yields the angular structure of I. This is done by plotting a point in the complex plane that is at the vector head of each phasor. For a given θ, two points located at the same radial distance from the origin but opposed by π in phase angle represent two wavelets that interfere destructively. Overlapping points represent constructive interference. As θ advances from zero the phasor-points rotate about the origin, causing the entire collection of points to evolve. By visually studying how the phasors move, one can see how regions in the particle progress from constructive to destructive interference and causes the rippled angular-structure of I.

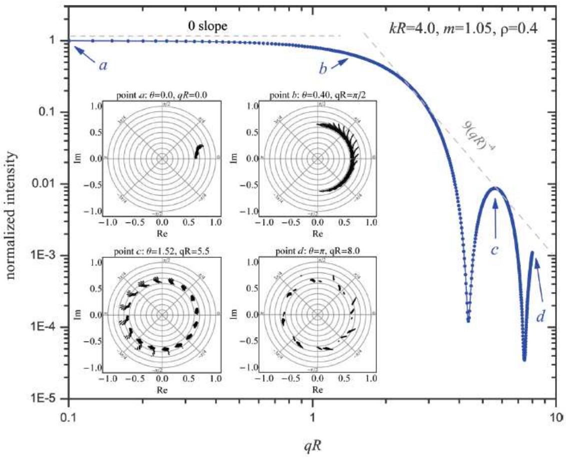

Figure 2. Phasor plots used to account for the power-law structure of I for a spherical particle with $kR = 4.0$ and $m = 1.05 + 0i$.

An example of this phasor analysis is presented in Fig. 2 where I is shown along with four phasor plots. This curve displays only two of the power law regions in Eq. (1) since $\rho < 1$. The phasor plots correspond to the points on I labeled $a - d$. One can see that the decrease in I at b corresponds to the spreading of the phasors as θ increases from zero, and hence the decrease in I is caused by destructive wavelet-interference. Further study reveals that the destructively interfering wavelets are those located at the extreme ends of the particle along the direction $\mathbf{q} = k(\hat{\mathbf{n}}^{\text{inc}} - \hat{\mathbf{r}})$, where $k\hat{\mathbf{n}}^{\text{inc}}$ is the incident wavevector. Thus, the power-law crossover at b is a signature of R.

Most particles have scattering curves that are far more complicated than the one in Fig. 2. An example is the particle in Fig. 3, where kR and m are greater than in Fig. 2, and I now displays all three power law regions of Eq. (1) along with both crossovers. As before, phasor plots are shown along with their corresponding points on I, except here the enhanced variation in magnitude and phase of $\mathbf{E}^{\text{int}}$ throughout the particle requires a more sophisticated analysis. Each phasor point is now assigned a color and brightness, where the color denotes the phasor's phase-angle when $\theta = 0$, i.e., in the forward-scattering direction, and the brightness denotes its magnitude. As the scattering angle is increased, and thus the phasors rotate in the complex plane, they *are not* recolored. This then allows one can track the evolution of each phasor by following its color as θ changes.

Phasor plot a in Fig. 3 corresponds to the forward-scattering direction from which the color and brightness of the phasors is assigned. As θ advances to point b, the rotation of the phasors causes some of the same color to become opposed by π indicating destructive interference between the corresponding wavelets. As before, these wavelets are associated with the inner regions in the particle maximally separated from each other along the $\mathbf{q}$ direction. This will be more explicitly described in the presentation.

Perhaps the more interesting feature in Fig. 3 is the ρ-crossover, which occurs approximately at point e. To understand its origin, refer to phasor plots b and e paying special attention to the largest-magnitude phasors, which are colored green. In b, these phasors are tightly clustered indicating that their wavelets contribute constructively to I at this scattering angle. However, in e these phasors have spread by approximately π in phase indicating that the wavelets associated with these green phasors are now destructively interfering. Consequently, the "extra" destructive interference beginning at point e accounts for the enhanced decrease in I at the ρ-crossover. The wavelets belonging to these green phasors reside in so-called hot-spots in the particle that are caused by refraction and lens-like effects of the particle-surface. The length scale associated with the ρ-crossover is then the approximate size of the hot spots.

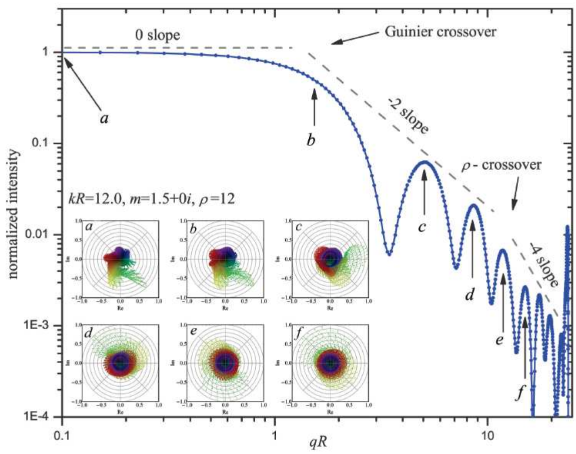

Figure 3. Scattered intensity for a spherical particle with $kR = 12$ and $m = 1.5 + 0i$ and corresponding color-coded phasor plots.

In special circumstances, the crossovers can have practical utility. For a single uniform particle, or a sufficiently dilute system consisting of many nearly identical spherical particles, an estimate for R can be obtained from the Guinier crossover in the measured intensity curve, e.g. see [4]. The quality of the estimate would rely on the fit of I to the power-laws defining the crossover. Note that this size estimate is largely independent of m. In addition, if I displays the ρ-crossover, then one also has an estimate for Re$\{m\}$, which comes from the combination of the Guinier-crossover R-estimate with $\rho = 2kR\text{Re}\{m-1\}$. Other practical concerns and the application of phasor analysis to nonspherical particles will be presented.

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