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# Rayleigh-Debye scattering with focused laser beams

Lee W. Casperson and C. Yeh

A focused beam technique has been developed for diagnosing the characteristics of individual particles within a polydisperse ensemble. In the Rayleigh-Debye approximation the scattered fields are related to the orientation and properties of a scatterer by means of explicit analytic formulas. The results simplify when the particle size is small compared to the minimum beam diameter.

## I. Introduction

Light scattering techniques have for many years provided an effective means for determining the concentration and properties of small particles suspended in a homogeneous medium. These techniques are much less useful, however, when the particle ensemble is polydisperse, i.e., the properties vary from particle to particle. With polydisperse media it has been necessary to physically isolate individual particles from the ensemble prior to applying an electromagnetic wave. Recently, however, we have described a new scattering technique in which the light source is a focused laser beam.<sup>1</sup> With typical scattering media, at most a single particle is likely to be near the focal region at any instant of time. Therefore, the radiation scattered from the focused beam may be used to deduce the properties of individual particles within an ensemble, and there is no need to remove the particles from their normal surroundings. In order to take full advantage of the focused beam technique, however, the relationships between the scattered fields and the particle properties must be known in detail. The purpose of this work is to develop realistic models for the scattering process which take proper account of the focused beam fields.

In the vast majority of treatments of electromagnetic scattering it is assumed that the unperturbed incident fields are homogeneous plane waves. If the scattering particles are spheres, an exact solution for this case is now commonly known as Lorentz-Mie or Mie scattering.<sup>2</sup> The Lorentz-Mie solutions and other exact results are numerically complex,<sup>3</sup> and several approximation methods have also been developed. The most generally

useful approximation is known as Rayleigh-Debye scattering.<sup>4</sup> In this approximation the additional phase shift experienced by a wave traversing the particle is required to be small compared to  $\pi$ . Thus the conditions of Rayleigh-Debye scattering are met if either the particle is small or its complex refractive index is close to unity. Following the work of Rayleigh and Debye this model was also discussed by Gans<sup>5</sup> and Rocard<sup>6</sup> and often goes by the name of Rayleigh-Gans or Rayleigh-Gans-Rocard scattering. The advantage of this method is that one can usually write simple explicit formulas for the scattered fields, even for nonspherical particles. Because of their simplicity such results are commonly applied when the basic phase shift condition is not well satisfied, and it is a remarkable fact that qualitatively useful results are sometimes still obtainable.

The subject of the present study is the application of the Rayleigh-Debye method to the problem of scattering from a focused Gaussian laser beam. In spite of the common usage of Gaussian beams in laboratory research, there have been very few treatments of light scattering by such beams. Pearson *et al.* have considered the scattering of plane wave Gaussian beams from edges.<sup>7</sup> Morita *et al.* have treated the scattering of a plane-wave beam from a small spherical particle at the beam axis.<sup>8</sup> Tsai and Pogorzelski generalized these results for an arbitrary sized spherical particle at the beam axis.<sup>9</sup> Recently, Tam has outlined a possible procedure for treating the scattering of a plane-wave beam from an off-axis sphere.<sup>10</sup> All these analyses have concerned the scattering of a plane wave having a Gaussian amplitude variation in the radial direction. Such a beam mode cannot be an exact solution of Maxwell's equations, and radial phase variations are always present except at the beam waist. A linearly polarized beam is also not possible in a rigorous solution, and minor field components must always be present in the longitudinal direction and in the transverse direction normal to the principal component. These extra field components are of no significance for most laser

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applications. However, with focused-beam scattering spatial resolution is best if the beam is brought to a very sharp focus with low  $f$ -number optics. Under these conditions the other field components, especially the longitudinal components, become comparable in magnitude to the principal transverse polarization components. A consideration of these other fields is thus essential for our present investigations. Recently the scattering of uniform converging beams by spheres has also been discussed.<sup>11</sup>

In Sec. II the basic scattering formulas for the Rayleigh-Debye model are derived in a form suitable for an arbitrary spatially dependent incident field and an arbitrarily shaped refracting particle. The field components of a focused Gaussian beam are derived in Sec. III, and general numerical solutions are presented. In Sec. IV a simplified central field model is introduced and discussed. The advantage of this model is that it leads to explicit analytic formulas for the scattered fields.

## II. Scattering Formulas

The essential feature of the Rayleigh-Debye approximation is that the electromagnetic field experienced by each volume element in the scatterer is the same as the incident field that would exist in the absence of the scatterer. The scattered field at any point in space is then a coherent superposition of the dipole radiation fields arising from each volume element. The result of this superposition can be inferred from the formulas for a phased array of conventional dipole antennas. This analogy forms the basis for the following discussion.

The complex amplitude of the retarded vector potential corresponding to a small antenna segment can be written<sup>12</sup>

$$A_z = \frac{\mu I dz}{4\pi\rho} \exp(-ik\rho) \quad (1)$$

where  $I$  is the peak value of the time-harmonic current flowing in the  $z$ -directed antenna segment of differential length  $dz$ . The coefficient  $\mu$  is the permeability of the medium surrounding the antenna,  $\rho$  is the distance between the antenna and the detection point, and  $k$  is the propagation constant. This antenna segment is basically a small dipole, and the dipole moment can be related to the current by<sup>13</sup>

$$p_z = -iI dz / \omega. \quad (2)$$

Combining these formulas, one finds that the vector potential of the scattered fields is related to the dipole moment  $\bar{p}$  by

$$\bar{A}_s = \frac{i\omega\mu\bar{p}}{4\pi\rho} \exp(-ik\rho). \quad (3)$$

If the dipole source is distributed in space, Eq. (3) must be replaced by

$$\bar{A}_s = \frac{i\omega\mu}{4\pi} \int_v \frac{\bar{p}(\bar{r})}{\rho} \exp(-ik\rho) dv, \quad (4)$$

where  $dv$  is a volume element in the scatterer.

To complete this formulation, the incident field may be related to the dipole moment, and the scattered field may be related to the vector potential. First, the dipole moment of a small dielectric body is given by<sup>14</sup>

$$\bar{p}(\bar{r}) dv = 3\epsilon_2 \left( \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + \epsilon_2} \right) \bar{E}(\bar{r}) dv, \quad (5)$$

where  $\epsilon_1$  is the permittivity of the scatterer,  $\epsilon_2$  is the permittivity of the surrounding medium, and  $\bar{E}(\bar{r})$  is the electric field that would have existed in the absence of the scatterer. Combining Eqs. (4) and (5), the vector potential of the scattered fields is

$$\bar{A}_s = \frac{3ik^2}{4\pi\omega} \left( \frac{n_1^2 - n_2^2}{n_1^2 + 2n_2^2} \right) \int_v \frac{\bar{E}(\bar{r}) \exp(-ik\rho)}{\rho} dv. \quad (6)$$

In this expression the propagation constant  $k$  is related to the permittivity and vacuum wavelength by

$$k = \omega(\mu\epsilon_2)^{1/2} = (2\pi n_2)/\lambda, \quad (7)$$

and the index of refraction is related to the permittivity by  $n_2 = (\epsilon_2/\epsilon_0)^{1/2}$ .

The scattered electric field amplitude at a point  $\bar{r}'$  is related to the vector potential by the equation<sup>15</sup>

$$\bar{E}_s(\bar{r}') = -i\omega \left\{ \bar{A}_s(\bar{r}') + \frac{1}{k^2} \nabla' [\nabla' \cdot \bar{A}_s(\bar{r}')] \right\}, \quad (8)$$

where  $\nabla'$  represents the gradient with respect to the primed coordinates. The total electric field at the point  $\bar{r}'$  may be obtained by adding on the field of the input beam according to

$$\bar{E}_t(\bar{r}') = \bar{E}_s(\bar{r}') + \bar{E}(\bar{r}'). \quad (9)$$

Equations (4)–(9) may be combined to yield the electric field at any point in space, and the result is

$$\begin{aligned} \bar{E}_t(\bar{r}') = & \frac{3k^2}{4\pi} \left( \frac{n_1^2 - n_2^2}{n_1^2 + 2n_2^2} \right) \int_v \left[ 1 + \frac{\nabla' \cdot (\nabla' \cdot)}{k^2} \right] \\ & \times \left[ \frac{\bar{E}(\bar{r})}{|\bar{r}' - \bar{r}|} \exp(-ik|\bar{r}' - \bar{r}|) \right] dv + \bar{E}(\bar{r}'), \end{aligned} \quad (10)$$

where the identity  $\rho = |\bar{r}' - \bar{r}|$  has been used. By employing the total field at a point inside of the scatterer as the input field for a second iteration, the accuracy of the Rayleigh-Debye method can be increased.<sup>16</sup> For our present purposes, however, the first order solutions are adequate.

Equation (10) can be simplified if we make the reasonable assumption that  $\rho$  is always large compared with the wavelength  $\lambda/n_2$ . The result is

$$\begin{aligned} \bar{E}_t(\bar{r}') = & \frac{3k^2}{4\pi} \left( \frac{n_1^2 - n_2^2}{n_1^2 + 2n_2^2} \right) \int_v \left\{ \frac{\bar{E}(\bar{r})}{|\bar{r}' - \bar{r}|} - \frac{(\bar{r}' - \bar{r})[(\bar{r}' - \bar{r}) \cdot \bar{E}(\bar{r})]}{|\bar{r}' - \bar{r}|^3} \right\} \\ & \times \exp(-ik|\bar{r}' - \bar{r}|) dv + \bar{E}(\bar{r}'). \end{aligned} \quad (11)$$

For the problem of scattering from a small object one can also assume that the distance  $\rho$  remains large compared to a typical dimension or displacement of the object. More specifically we require that every point of the object be close to the coordinate reference so the following conditions apply:

$$\begin{aligned} x' \gg x, \quad y' \gg y, \quad z' \gg z, \\ \rho \approx (x'^2 + y'^2 + z'^2)^{1/2}. \end{aligned} \quad (12)$$

Now Eq. (11) reduces to

$$\bar{E}_t(\bar{r}') = \frac{3k^2}{4\pi} \left( \frac{n_1^2 - n_2^2}{n_1^2 + 2n_2^2} \right) \cdot \frac{\exp(-ik\rho)}{\rho} \left[ \bar{S} - \frac{\bar{r}'(\bar{r}' \cdot \bar{S})}{\rho^2} \right] + \bar{E}(\bar{r}'), \quad (13)$$

where  $\bar{S}$  represents the integral

$$\bar{S} = \int_v \bar{E}(\bar{r}) \exp(ik\bar{r} \cdot \bar{r}'/\rho) dv. \quad (14)$$

Equation (13) can also be written in the forms

$$E_t(\bar{r}') = -\frac{3k^2}{4\pi} \left( \frac{n_1^2 - n_2^2}{n_1^2 + 2n_2^2} \right) \frac{\exp(-ik\rho)}{\rho^3} [\bar{r}' \times (\bar{r} \times \bar{S})] + \bar{E}(\bar{r}') \quad (15)$$

$$= \frac{3k^2}{4\pi} \left( \frac{n_1^2 - n_2^2}{n_1^2 + 2n_2^2} \right) \frac{\exp(-ik\rho)}{\rho} (\sin\chi) |\bar{S}| \bar{i}_s + \bar{E}(\bar{r}'), \quad (16)$$

where  $\chi$  is the angle between the  $\bar{S}$  vector and  $\bar{r}'$ , and  $\bar{i}_s$  represents the polarization direction of the scattered field.

Equations (13) and (14) are our basic results for scattering with focused beams, and it is appropriate to compare these formulas with previous treatments. One significant feature here is that the spatial variations of the incident field prevent the easy removal of  $\bar{E}(\bar{r})$  from the integral in Eq. (14). If the field had been a homogeneous plane wave, this removal would have been possible, and Eq. (13) would simplify to well known formulas. Analytic techniques for reducing Eq. (14) with a Gaussian input field are discussed in Sec. IV. Another difference from some treatments is that the incident field also is a direct component of the total detected field, and this is due to the large beamwidth of the incident field.

### III. Focused Beam Fields

In order to apply the previously derived scattering formulas, explicit procedures are needed for obtaining the components of the incident electromagnetic wave. Unfortunately, there exist no exact analytic solutions of Maxwell's equations for freely propagating beams. Such solutions can only be obtained by numerical integration of the wave equation or by a Fourier expansion in terms of infinite plane waves.<sup>17</sup> It is also reasonable to enquire whether sufficient accuracy might be obtainable using approximate analytical expressions for the field components. The dominant transverse field components can be expressed in terms of polynomial-Gaussian functions.<sup>18</sup> In this section approximate formulas are given for the fundamental Gaussian beam including first-order corrections for nontransverse field components.<sup>19</sup> Detailed solutions are presented, and the significance of the correction terms is explored.

Maxwell's equations for the complex amplitudes of the electric and magnetic fields can be written in the well known form

$$\nabla \times \bar{E} = -i\omega\mu\bar{H}, \quad (17)$$

$$\nabla \times \bar{H} = i\omega\epsilon\bar{E}. \quad (18)$$

In a homogeneous medium these equations may be combined to yield the wave equation

$$\nabla^2 \bar{E} + k^2 \bar{E} = 0, \quad (19)$$

where the propagation constant is  $k = \omega(\mu\epsilon)^{1/2}$ . In approximate solutions of Eq. (19) one typically assumes that the beam is nearly a linearly polarized plane wave in the form

$$E_x = \psi(x,y,z) \exp(-ikz), \quad (20)$$

where  $\psi(x,y,z)$  is a slowly varying function of the spatial coordinates. When Eq. (20) is substituted into Eq. (19), one obtains the new wave equation

$$\frac{\partial^2 \psi}{\partial x^2} + \frac{\partial^2 \psi}{\partial y^2} + \frac{\partial^2 \psi}{\partial z^2} - 2ik \frac{\partial \psi}{\partial z} = 0. \quad (21)$$

In the paraxial approximation the second derivative of  $\psi$  with respect to  $z$  is neglected, and several complete orthogonal sets of solutions of the resulting equation are known. Of particular interest here is the fundamental Gaussian mode which may be shown by direct substitution to have the form

$$\psi = \psi_0 \exp[-iQ(z)(x^2 + y^2)/2 - iP(z)], \quad (22)$$

where the parameters  $Q$  and  $P$  have the  $z$  dependences

$$Q(z) = Q_0(1 + zQ_0/k)^{-1}, \quad (23)$$

$$P(z) = P_0 - i \ln(1 + zQ_0/k). \quad (24)$$

In these results  $Q_0$  and  $P_0$  represent, respectively, the values of  $Q(z)$  and  $P(z)$  at the beam waist ( $z = 0$ ).  $Q(z)$  can be identified with the phase front curvature  $R(z)$  and the  $1/e$  amplitude spotsize  $w(z)$  using the equation

$$Q(z) = k/R(z) - 2i/w^2(z), \quad (25)$$

and  $P(z)$  is the complex phase.

For the present application the beams are strongly focused, and there is no *a priori* reason for assuming that the paraxial approximation will provide a useful description of the focused beam fields. Therefore, we have employed an iterative procedure for obtaining increasingly accurate descriptions of the fields. This procedure involves substituting an approximate expression for the field components into the left-hand sides of Eqs. (17) and (18) in order to obtain new expressions for  $\bar{E}$  and  $\bar{H}$ . As our starting approximation we use the Gaussian beam solution given in Eq. (22). Thus a reasonable set of starting fields is

$$E_x(x,y,z) = \psi(x,y,z) \exp(-ikz), \quad (26)$$

$$H_y(x,y,z) = (\epsilon/\mu)^{1/2} \psi(x,y,z) \exp(-ikz). \quad (27)$$

When Eqs. (26) and (27) are introduced into the left-hand sides of Eqs. (17) and (18), the leading  $z$  components are found to be

$$E_z(x,y,z) = -xQ(z)\psi(x,y,z) \exp(-ikz)/k, \quad (28)$$

$$H_z(x,y,z) = -(\epsilon/\mu)^{1/2} yQ(z)\psi(x,y,z) \exp(-ikz)/k. \quad (29)$$

We may assume that  $x$  (or  $y$ ) is at most comparable to  $w$ , and near the beam waist  $w$  is on the order of  $|Q|^{-1/2}$ . Therefore, the  $z$  components are smaller than the leading transverse components by a factor of about  $(kw)^{-1}$ .

The orthogonal field components and other corrections can be found by further iterations. In establishing the magnitude of these corrections it suffices to assume that the most rapid  $z$  variations occur in the terms  $\exp(-ikz)$ . Thus self-consistent expressions for the orthogonal components are

$$E_y(x,y,z) = -xyQ^2(z)\psi(x,y,z) \exp(-ikz)/2k^2, \quad (30)$$

$$H_x(x,y,z) = -(\epsilon/u)^{1/2}xyQ^2(z)\psi(x,y,z) \exp(-ikz)/2k^2. \quad (31)$$

Near the beam waist the magnitude of these components is smaller than the dominant transverse components by about  $(kw)^{-2}$ . This is also the approximate amount by which the higher order corrections to  $E_x$  and  $H_y$  are smaller than the leading terms, and these corrections are not necessary for the present discussion. The smallness of the corrections is easily confirmed with a specific example. The smallest spot size that one might expect to encounter in practice is about  $w = \lambda$ , and the correction factor is already as small as  $(kw)^{-2} = 0.0025$ .

#### IV. Central Field Approximation

In Sec. II a general formula has been developed for calculating the scattered field distribution that results when an arbitrary electromagnetic beam is incident on a homogeneous particle. In essence the problem is reduced to evaluating the integral in Eq. (14). Unfortunately, this integral can be performed explicitly only for certain simple distributions of the electromagnetic fields. For the Gaussian field distributions of focused beams the integrals cannot be expressed in terms of elementary functions. Numerical integration is always straightforward, and some solutions will be discussed. However, it has proved to be very useful in our work to introduce a simpler model for the electromagnetic fields, and the result may be called a central field approximation. The basic geometry that we have in mind is illustrated in Fig. 1. The incident beam is a Gaussian having its waist at the plane  $z = 0$ .

Combining Eqs. (14), (26), and (28), we find that the general form for the scattering integral is

$$S_x = \int_v \psi(x,y,z) \exp(i\delta) dv, \quad (32)$$

$$S_z = - \int_v xQ(z)\psi(x,y,z) \exp(i\delta) dv, \quad (33)$$

where the term  $\delta = k(\vec{r} \cdot \vec{r}'/\rho - z)$  in the exponent represents the phase shift of a scattered ray with respect to a ray travelling straight from the origin. These integrals cannot be performed explicitly. However, if the particle size is small or at most comparable to the spot size, simplifications are possible. Such a smallness constraint may apply to many reasonable practical configurations and is consistent with the basic phase shift condition underlying the Rayleigh-Debye approximation. In this case the spatial variations of the

field in the vicinity of the particle can be accurately described using only a few terms in a power series expansion. The expansion is performed about some typical location in the particle, and for a spherical scatterer the natural expansion point is the particle center. Thus the Gaussian field distributions described in the previous section can be expanded in a Taylor series with respect to the Cartesian coordinates, and some of the integrals represented by Eq. (14) may be performed analytically.

In our own studies the general expansions have often proved to be unnecessarily accurate, and for brevity we only present the simplest limiting cases. The first step is to observe that for a particle small compared to the beam diameter the only  $z$  variation of importance in Eqs. (26) and (28) is the  $\exp(-ikz)$  plane wave factor. Thus, Eqs. (32) and (33) can be written

$$S_x = \int_x \int_y \psi(x,y,z_1) \int_z \exp(i\delta) dz dy dx, \quad (34)$$

$$S_z = - \int_x \int_y xQ(z_1)\psi(x,y,z_1) \int_z \exp(i\delta) dz dy dx, \quad (35)$$

where  $z_1$  is the coordinate of the center of the particle (or some other characteristic reference). With a change of coordinates the  $z$  integration reduces to the integration of a simple exponential.

A slightly less accurate subsequent approximation is to assume that the transverse field variations in the vicinity of the particle are entirely negligible. Thus, Eqs. (34) and (35) reduce to

$$S_x = \psi(x_1,y_1,z_1) \int_v \exp(i\delta) dv, \quad (36)$$

$$S_z = -x_1Q(z_1)\psi(x_1,y_1,z_1) \int_v \exp(i\delta) dv, \quad (37)$$

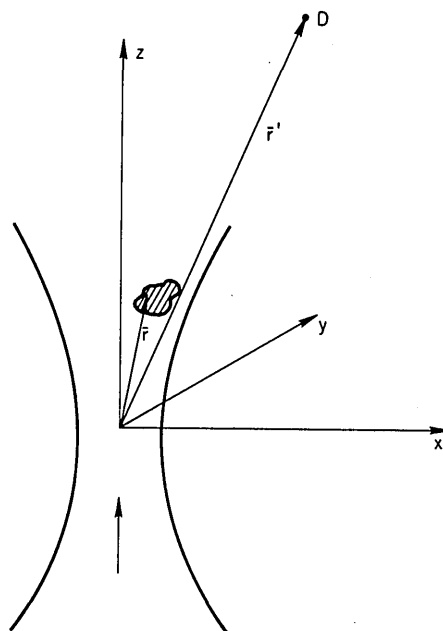


Fig. 1. Coordinate geometry used in the analysis of scattering with focused beams. The beam waist is at  $z = 0$ , and  $D$  represents a detection point.

where  $x_1$  and  $y_1$  are the transverse coordinates of the particle center. These remaining integrations can be performed explicitly for many simple particle shapes. In the case of spheres the result is<sup>20</sup>

$$\int_v \exp(i\delta) dv = vG(u), \quad (38)$$

where  $v$  is the volume of the sphere, and the variable  $u$  is related to the scattering angle  $\theta$  and the particle radius  $a$  by

$$u = 2ka \sin(\theta/2). \quad (39)$$

The function  $G(u)$  is given by

$$G(u) = 3v(\sin u - u \cos u)/u^3. \quad (40)$$

Thus, Eqs. (36) and (37) now have the explicit forms

$$S_x = v\psi(x_1, y_1, z_1)G(u), \quad (41)$$

$$S_z = -x_1 Q(z_1) v\psi(x_1, y_1, z_1)G(u). \quad (42)$$

The preceding equations and their obvious extensions to higher orders constitute the essence of the central field approximation. In the simplest limit the field is assumed to be a constant in the vicinity of the particle, and the value of this constant is the actual value of the field at the center of the particle.

It is now reasonable to inquire whether these equations reduce in the appropriate limit to the well known expressions for Rayleigh-Debye scattering of homogeneous plane waves. A Gaussian beam reduces to a plane wave if the spot size  $w$  and phase front curvature  $R$  are permitted to increase without limit. Therefore, from Eq. (25) the beam parameter  $Q$  vanishes and with it any longitudinal or orthogonal field components implied by Eqs. (28)–(31). Equation (13) now reduces to

$$\begin{aligned} \bar{E}_t(r') = \frac{3k^2 v \psi}{4\pi} \left( \frac{n_1^2 - n_2^2}{n_1^2 + 2n_2^2} \right) G(u) \frac{\exp(-ik\rho)}{\rho} \\ \times \left[ \bar{i}_x - \frac{\bar{r}'(\bar{r}' \cdot \bar{i}_x)}{\rho^2} \right] + \bar{E}(r'). \end{aligned} \quad (43)$$

The magnitude of the bracketed terms can sometimes be replaced by unity or  $\cos\theta$  if the incident radiation is polarized, respectively, perpendicular to the scattering plane or parallel to the scattering plane. These results are in agreement with the conventional Rayleigh-Debye scattering formulas.<sup>4</sup>

An important application of the scattering formalism that has been developed is the determination of the location and properties of small scatterers. For this purpose one would like to be able to obtain a unique inversion of the formulas, and it is desirable to know whether such an inversion is possible. This question can be most easily answered by considering the symmetry characteristics of the scattering formulas themselves. It is assumed that the direct beam  $\bar{E}(\bar{r}')$  has been eliminated from the scattering pattern represented by Eq. (13), and this term will be included in the later discussion.

We treat first the case in which the scattering particle is inverted across the  $z$  axis in such a way that the dielectric constant at every point  $(x, y, z)$  is replaced by the

dielectric constant of the corresponding point  $(-x, -y, z)$ . Simultaneously the detection point  $(x', y', z')$  is moved to the point  $(-x', -y', z')$ . From Eqs. (32) and (33) and the symmetry of the beam it follows that this transformation leaves  $S_x$  (and  $S_y$ ) unchanged while the sign of  $S_z$  is reversed. Now an examination of Eq. (13) reveals that  $E_{tx}$  and  $E_{ty}$  are unchanged, while the sign of  $E_{tz}$  is reversed. This last sign change has no effect on the energy density  $u_e \propto \bar{E} \cdot \bar{E}^*$ , so the measured intensity is not altered by the transformation. This result is reasonable, since we expect intuitively that a reversal of scatterer and detector should yield no change in the output.

Somewhat more subtle is the question of what happens if the detector is inverted across the  $z$  axis while the scatterer is inverted across the  $z = 0$  plane. Thus we replace the dielectric constant at every point  $(x, y, z)$  by the dielectric constant at the opposite point  $(x, y, -z)$ , and the detection point  $(x', y', z')$  is replaced by the point  $(-x', -y', z')$ . From Eqs. (23) and (24) the real parts of  $Q(z)$  and  $P(z)$  change sign (with  $P_0 = 0$ ), and the imaginary parts are unchanged. Then from Eq. (22) (with  $\psi_0$  real)  $\psi$  is replaced by its complex conjugate. Equations (26)–(31) imply that  $E_x$  and  $E_y$  are replaced by their complex conjugates while  $E_z$  is replaced by its negative conjugate. Next, Eqs. (32) and (33) imply that  $S_x$  (and  $S_y$ ) are replaced by their conjugates while  $S_z$  is replaced by its negative conjugate. Finally, with Eq. (13) one finds again that the observed intensity is unchanged by the transformation.

Combining the previously derived symmetry relations (or performing a third derivation) one discovers that there is a basic degeneracy in the scattering formulas. In particular, the field distribution is unchanged in this Rayleigh-Debye approximation when the scatterer is inverted across the origin  $(0, 0, 0)$ . This degeneracy is an unfortunate source of uncertainty unless either the particle shape or its location can be determined by some independent means. If the particles themselves possess inversion symmetry, the degeneracy makes the scattering problem nonunique. For example, the scattering pattern of a sphere centered at the point  $(x_1, y_1, z_1)$  is exactly the same as the pattern for the sphere located at the point  $(-x_1, -y_1, -z_1)$ . Furthermore, a sphere situated anywhere on the  $x$  axis somewhat surprisingly yields a scattering pattern which is symmetric in the detection coordinate  $x'$ .

An examination of Eq. (13) shows that the degeneracy is removed if the incident field  $\bar{E}(\bar{r}')$  is included in the detected radiation. Then the scattering pattern is superimposed with the intense forward beam, and a characteristic pattern of interference fringes occurs where the two signals are comparable in amplitude. The lack of uniqueness can also be overcome by additional complexity in the experimental apparatus. For example, if the focal point of the incident beam is dithered in any direction on a time scale short compared to the transit time of the particle, the response of the scattered fields will eliminate the uniqueness problem. Alternatively, spatial filtering in front of the detectors can reveal additional information about the particle

location. To eliminate the degeneracy it would be sufficient, for example, to know whether the particle is above or below the  $z = 0$  plane.

## V. Results

The implications of the scattering formulas that have been obtained can be most readily illustrated by means of specific examples. The examples chosen correspond to the configurations of certain of our experiments. The assumed radiation wavelength is  $\lambda = 0.5145 \times 10^{-6}$  m corresponding to the blue-green line of an argon laser. The scattering particles are latex spheres of  $0.24 \times 10^{-6}$  m radius and refractive index  $n_1 = 1.59$ . These spheres are suspended in water with a refractive index of  $n_2 = 1.33$ , and the beam is focused in the water to a minimum spot size of  $w_0 = 0.54 \times 10^{-6}$  m. The purpose of these calculations is to check on the accuracy of the central field approximation and to see how the scattered intensity depends on the location of the scatterer.

The scattered field distribution in the  $z' = 1$ -cm plane is plotted in Fig. 2 for the spherical particle at two locations in the  $z = 0$  plane. The incident field is polarized primarily in the  $x$  direction, and the normalized energy density is  $u = \vec{E}_t \cdot \vec{E}_t^*$  with  $\psi_0 = 1$ . The solid lines are the general Rayleigh-Debye results based upon Eqs. (13), (32), and (33). The incident field  $\vec{E}(\vec{r}')$  is omitted from Eq. (13) as discussed previously. The dashed lines are the central field approximation of Eqs. (13), (41), and (42). It is apparent from the figure that even in this extreme case of a particle comparable in size to the beam the central field model is accurate within a few percent. The central field model exaggerates the scattering in

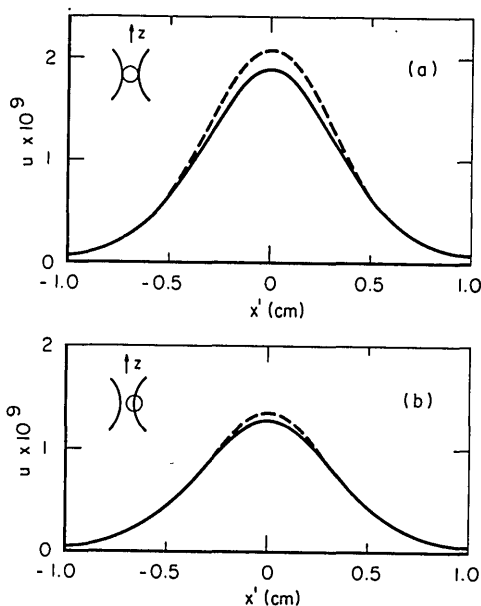


Fig. 2. Scattered field distribution along the  $x'$  axis in the  $z = 1$ -cm plane for (a) a particle at the origin  $x = 0$  and (b) a particle at  $x = 0.25 \times 10^{-6}$  m. The solid lines represent the exact Rayleigh-Debye solutions, and the dashed lines are the central field results. The particle and beam properties are described in the text. Note that the scattered field remains symmetric in  $x'$  even though the particle in (b) is off axis.

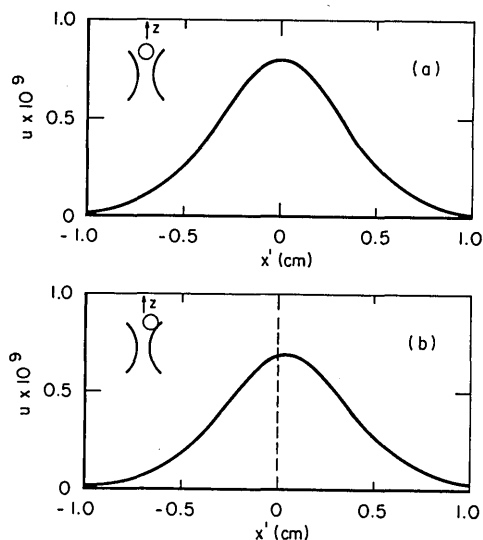


Fig. 3. Scattered field distribution along the  $x'$  axis in the  $z = 1$ -cm plane for (a) a particle at the coordinate  $x = 0, y = 0, z = 3 \times 10^{-6}$  m and (b) a particle at the coordinates  $x = 0.25 \times 10^{-6}$  m,  $y = 0, z = 3 \times 10^{-6}$  m. Note that in case (b) the scattered fields are slightly asymmetric in  $x'$ .

Fig. 2(a) because it also exaggerates the incident field near the edge of the particle. The symmetry of the scattering pattern in Fig. 2(b) follows from the relations of the previous section.

In Fig. 3 are presented the corresponding results for a scattering particle located in the  $z = 3 \times 10^{-6}$ -m plane. The scattered intensity is reduced because the particle is away from the focus. A small but distinct asymmetry always appears when the particle is away from the  $z$  axis (except in the  $z = 0$  plane). The agreement between the exact and central field models in this case is too close to be represented in the figure, and both models imply the asymmetry for off-axis particles. This slight deflection of the scattered radiation beam can be qualitatively understood from a consideration of the wavefront orientation at the particle.

## VI. Conclusion

A Rayleigh-Debye method has been developed for treating the scattering of focused light beams, and this is the first focused beam treatment which accounts for nontransverse components of the electromagnetic fields. Any focused beam must possess such nontransverse components, and the examples considered here have involved the important Gaussian beams which commonly result with laser sources. Another area of investigation has involved reducing the volume integrals inherent in the Rayleigh-Debye method to explicit analytic formulas. This has been accomplished by a sequence of central field approximations. The result of this study is that even the poorest of these approximations retains the major field asymmetries and differs by at most a few percent from the general Rayleigh-Debye values. It is also worth noting that the general shape of the scattering diagrams is largely insensitive to the

position of the particle in the beam and differ in relatively minor respects from the scattering patterns for homogeneous plane waves. This fact should lead to a considerable simplification of the interpretation and inversion of focused beam scattering data.

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