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Acoustic scattering by a spherical obstacle: Modification to the analytical long-wavelength solution for the zero-order coefficient

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Classical long wavelength approximate solutions to the scattering of acoustic waves by a spherical liquid particle suspended in a liquid (an emulsion) show small but significant differences from full solutions at very low $k_c a$ (typically $k_c a < 0.01$) and above at $k_c a > 0.1$, where k_c is the compressional wavenumber and a the particle radius. These differences may be significant in the context of dispersed particle size estimates based on compression wave attenuation measurements. This paper gives an explanation of how these differences arise from approximations based on the significance of terms in the *modulus* of the complex zero-order partial wave coefficient, A_0 . It is proposed that a more accurate approximation results from considering the terms in the real and imaginary parts of the coefficient, separately. © 2011 Acoustical Society of America. [DOI: 10.1121/1.3543967]

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I. INTRODUCTION

The scattering of sound waves by a spherical obstacle is a fundamental physical problem which impacts on the use of acoustic and ultrasonic techniques in a wide variety of applications. Indeed, ultrasonic spectroscopy has found application in industries as diverse as pharma, food, health care, and aerospace. Many such techniques rely on the interpretation of the measured sound speed or attenuation in terms of scattering by individual components, whether particles, droplets, molecular aggregates, or cavities, all of which are often approximated as spheres. Hence, the scattering of a planar sound wave by a sphere forms the basis of many ultrasonic techniques. In the present work, it is the long wavelength region which is of interest, where the wavelength of the compressional wave is much larger than the radius of the sphere. This region is pertinent to many real systems, using ultrasonic frequencies (1–50 MHz) with the colloidal (micron-scale) particles found in many stable emulsions or suspensions, or micron-sized cavities.

The study of the spherical scatterer problem has a long history, with the work of Rayleigh in the 19th century¹ still forming the basis of the scattering model in use currently. The principles of the Rayleigh partial wave method were applied to liquid in liquid, solid in liquid, and solid in solid systems (emulsions, suspensions, and solid systems) by Epstein and Carhart,² Allegra and Hawley,³ and Ying and Truell,⁴ respectively, extending the model to account for thermal and shear processes. Their models for scattering by an isolated sphere have been shown to be the special cases of a single generalized model applicable to any phase (liquid, solid, gas) for the sphere or the surrounding medium, now known as the ECAH model.⁵ When combined with an appropriate multiple scattering theory to account for scatter-

ing by many particles, these models have been adopted for interpretation of ultrasonic measurements. A review of the models and their applications can be found in Challis *et al.*⁶

Although it provides a solution to the scattering problem, the ECAH formulation, as it has become known, can be numerically challenging, relying on inversion of complex matrices which are ill-conditioned. As a result, several workers have attempted to provide analytical solutions for the scattering coefficients, especially in the very low frequency, long wavelength region where only two or three of the coefficients are relevant. Indeed, the original workers provided analytical approximations for the low frequency limits.^{2–4} These have the advantage of numerical simplicity and stability, and lend themselves to fast and simple computation which might be required, for example, in a real-time measurement system. Using a different formulation, Isakovich derived the wavenumber for a dilute liquid-in-liquid system of particles, accounting for the sound absorption due to thermal dissipation.⁷ His result can be shown to be almost identical with the ECAH formulation when written in comparable form.^{8,9} In more recent times, other researchers have developed an alternative formulation for the sphere scattering problem which evaluates the scattered field as a series in the wavenumber-radius product, $k_c a$, thus permitting calculation at low frequency without matrix inversion.^{10–14} In addition, an analytical solution for the lowest order coefficients was derived by one of the present authors, which is not restricted to low frequencies, but which does retain some complex functions of the wavenumber-radius products.¹⁵ The early analytical solutions for the scattering coefficients, both from the ECAH formulation³ and from the Isakovich formulation⁷ have also been adopted in studies which have incorporated further physical effects into the model, such as the thermal overlap which occurs in relatively concentrated emulsions.^{8,9,16}

The analytical or “explicit” solution to the scattering coefficients in the ECAH formulation has some application

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for interpretation of ultrasound measurements in the long wavelength (low frequency) region. However, we have identified that the solution for the zero-order coefficient deviates from the full solution under some conditions even in this region by as much as 100% on attenuation, notably for very small particle radii, where it would be expected to be accurate. We believe that this may be a result of the neglect of terms in the expansions of the functions in the scattering coefficients used to obtain the analytical low-frequency forms. We report here the derivations of new expansions of the analytical results for the zero-order scattering coefficient, for a liquid-in-liquid spherical scatterer, retaining all significant terms. Numerical calculations are presented to demonstrate the accuracy of the new results.

II. THE ECAH METHOD

Our analysis is based on the ECAH formulation of the Rayleigh partial-wave method for scattering by a spherical fluid object in a fluid medium. Details can be found in a recent review article,⁶ or in the original papers.^{2,3} Here, only a brief summary is given. The scattering at the particle produces three wave modes in both the particle and the surrounding fluid: two longitudinal modes (propagational and thermal) and one shear mode. The wavenumbers, k_c for the propagational mode, k_T for the thermal mode, and k_S for the shear mode, are given to a very good approximation at angular frequency ω in fluids by

$$\begin{aligned} k_c &= \frac{\omega}{c} + i\alpha & k_T &= \left(\frac{\omega}{2\sigma}\right)^{1/2} (1+i), \\ k_S &= \left(\frac{\omega}{2\nu}\right)^{1/2} (1+i), \end{aligned} \quad (1)$$

where c is the speed of sound, α is the attenuation, σ is the thermal diffusivity; such that $\sigma = \tau/\rho C_p$, where τ is the thermal conductivity, ρ is the density, and C_p is the specific heat capacity at constant pressure; and $\nu = \eta/\rho$ is the kinematic viscosity, where η is the shear viscosity. In the notation used in this paper, primed quantities refer to the inside of the particle, and subscripts c , T , and S refer to the propagational, thermal, and shear modes, respectively.

The scattered, outgoing, compressional wave resulting from a planar compressional wave incident on the sphere, is written in the form of a scalar velocity potential, φ , expressed as a sum of partial wave modes thus

$$\varphi = \sum_{n=0}^{\infty} i^n (2n+1) A_n h_n(k_c a) P_n(\cos \theta), \quad (2)$$

where n is the integer order of the partial wave mode, $h_n(x)$ is the spherical Hankel function, $P_n(\cos \theta)$ are Legendre polynomials, θ is the polar angle, k_c is the compressional wavenumber of the medium surrounding the spherical scatterer, and a is the radius. A time dependence of the form of $e^{-i\omega t}$ is assumed. The scattering coefficients A_n are the amplitudes of the partial wave modes and are determined by the boundary conditions at the surface of the particle.

In the long wavelength region, only the lowest order partial wave modes are significant, and it is common in this

region to use only the zero and first order coefficients. The A_0 coefficient characterises the breathing motion of the scatterer which is affected both by the compressibility difference between the scatterer and the surrounding medium, and the difference in the thermal properties of the two media. The first order coefficient A_1 results from inertial effects due to density difference and from the viscous drag of the surrounding fluid.

In an emulsion, the combination of the scattered fields can be expressed as a wavenumber for the emulsion, $K = \omega/c_{\text{sol}} + i\alpha_{\text{sol}}$, where c_{sol} is the speed, and α_{sol} is the attenuation of sound propagation through the emulsion. Using the multiple scattering theory of Lloyd and Berry,¹⁷ the wavenumber can be related to the scattering coefficients A_n by

$$\begin{aligned} \left(\frac{K}{k_c}\right)^2 &= 1 + \frac{3\phi}{k_c^2 a^3} f(0) + \frac{9\phi^2}{4k_c^4 a^6} \left(f^2(\pi) - f^2(0) \right. \\ &\quad \left. - \int_0^\pi d\theta \frac{1}{\sin(\theta/2)} \left(\frac{d}{d\theta} f^2(\theta) \right) \right), \end{aligned} \quad (3)$$

where the far-field scattering amplitude is defined by

$$f(\theta) = \frac{1}{ik_c} \sum_{n=0}^{\infty} (2n+1) A_n P_n(\cos \theta). \quad (4)$$

Note that here the symbol ϕ refers to the volume fraction of the dispersed particles. The Lloyd and Berry formula has been successfully applied to acoustic measurements for many years,⁶ although it does neglect the (usually) small contribution from the multiple scattering of the thermal and shear scattered wave terms. Various cell-type models have been constructed to account for these additional effects; but at relatively low concentrations, Eq. (3) has found widespread application.⁶

Thus in the long wavelength region, where only the zero and first order coefficients are significant, and in a dilute system

$$\left(\frac{K}{k_c}\right)^2 \approx 1 - \frac{3\phi i}{k_c^3 a^3} (A_0 + 3A_1). \quad (5)$$

In the work reported here, a new analytical form of the zero order coefficient A_0 in the ECAH formulation is derived which is valid at a low frequency (i.e., the long wavelength region).

III. MODIFIED COEFFICIENT A_0

In order to obtain an analytical solution for A_0 at low frequency, where $|k_c a| \ll 1$, which avoids the use of complex functions such as the Hankel and Bessel functions, the solution from the boundary conditions is expressed as a series in the wavenumber radius product $k_c a$, retaining only the first or a small number of terms. It should be noted that the thermal and shear wavenumber-radius products (e.g., $k_T a$) cannot be assumed to be small, but that $|k_c/k_T|^2 \ll 1$. In addition, $|k'_c a|$ is assumed to be of similar magnitude to $|k_c a|$.

Our expansion is based on the analytical solution for A_0 derived by Pinfield¹⁵ [Eqs. (55–56) and (61–62)], which is

not restricted to small $k_c a$ and is conveniently separated into non-thermal and thermal parts. For each part, the numerator and denominator were each expressed as a series expansion in $k_c a$. Whereas previous workers neglected terms of order $(k_c a)^2$ relative to unity, we have retained these terms where they make a significant contribution to either the real or imaginary part of the expression. Since the attenuation predominantly relates to the real part of the scattering coefficient, and speed to the imaginary part, it is crucial that both real and imaginary parts are sufficiently accurate independently. The absolute value of the coefficient is dominated by its imaginary part (relating to speed). Thus, terms which are not significant in the absolute value of the coefficient may still make a significant, if not dominant, contribution to the real part of the coefficient. It is believed that the neglect of such terms in the original derivations by ECAH may have led to the inaccuracies observed. It should be noted that both k_T^2 and k_S^2 are imaginary for fluids, so that terms including these parameters are likely to contribute to previously neglected but significant components of the expression. In addition it was found by numerical calculations that some higher order terms were significant under certain conditions. Algebraic developments were carried out using MAPLE software.¹⁸

The zero-order scattering coefficient, A_0 , is expressed as a sum of a non-thermal, A_{0N} , and a thermal, A_{0T} , component as follows:

$$A_0 = A_{0N} + A_{0T}. \quad (6)$$

For the non-thermal part of the coefficient, the derivation is commenced from Eq. (56) of Ref. 15, taking series expansions of the numerator and denominator independently and neglecting terms of order $(k_c a)^3$ and higher, which are small relative to unity. In the numerator, the dominant term is of order $(k_c a)^2$ and is predominantly real. However the largest imaginary component of the numerator appears in the term of order $(k_c a)^4$; this term would have been neglected if terms of order $(k_c a)^2$ (relative to unity) had been neglected without considering their real or imaginary character. However, we have neglected *real* terms of order $(k_c a)^4$. Thus the numerator is approximated by

$$A_{0N\text{numer}} = -\frac{\hat{\rho}}{3} \left[1 - \frac{(k'_c a)^2}{(k_c a)^2 \hat{\rho}} \right] (k_c a)^2 + \frac{4(k'_c a)^2 (\hat{\eta} - 1)}{9(k_c a)^2 (k_s a)^2} (k_c a)^4. \quad (7)$$

The circumflex is used to denote the ratio of the physical property in the dispersed phase (inside the particle) and the surrounding medium, so $\hat{\rho} = \rho'/\rho$. In the denominator, the most significant contribution is imaginary and of order $(k_c a)^{-1}$, but by the same process, the dominant real contribution was identified with order $(k_c a)$. Thus the denominator can be approximated by

$$A_{0N\text{denom}} = -\frac{i\hat{\rho}}{(k_c a)} + \frac{4i(k'_c a)^2 (\hat{\eta} - 1)}{3(k_c a)^2 (k_s a)^2} (k_c a), \quad (8)$$

neglecting the imaginary terms of order $(k_c a)$. Taking the binomial expansion of the denominator to obtain the approximated solution for A_{0N} leads to

$$A_{0N} = -\frac{i(k_c a)^3}{3} \left[1 - \frac{(k'_c a)^2}{(k_c a)^2 \hat{\rho}} \right] \left[1 + \frac{4(k'_c a)^2 (\hat{\eta} - 1)}{3\hat{\rho} (k_s a)^2} \right]. \quad (9)$$

The second square bracket is a new term in the expansion, as seen when compared with the previous solution for the non-thermal term, which can be found in the original works [Ref. 2 Eq. (10.2) and Ref. 3 Eq. (13)] and is also quoted by McClements *et al.*,⁹ thus:

$$A_{0N} = -\frac{i(k_c a)^3}{3} \left[1 - \frac{(k'_c a)^2}{(k_c a)^2 \hat{\rho}} \right]. \quad (10)$$

The new term in the non-thermal part of the coefficient relates to the ratio $(k'_c a)^2 / (k_s a)^2$ which, although small in magnitude, is largely imaginary. Thus it contributes to the dominant real part of the coefficient under some conditions, since the other contributions result from the (small) imaginary parts of the wavenumber k_c . Thus, the neglect of this new correction leads to inaccuracies in the real part of the coefficient, which affects the attenuation.

The thermal term has a considerably more complicated result. Derivation begins with Eq. (61) of Ref. 15; for the ease of manipulation, both numerator and denominator were written such as to avoid quotients in each. Thus the denominators of the A_{0N} and A'_{0N} components appear in the denominator of A_{0T} . In order to identify the dominant real and imaginary parts, we first examined the complex character of the component parameters such as the thermal factors and the functions of the thermal wavenumber, b_1 – b_3 [Eqs. (52)–(54), Ref. 15]. For liquids (where a velocity potential is used), the factors relating the wave potential to the temperature for the compressional and thermal modes, respectively, are³

$$\Gamma_c = -\frac{\gamma}{(-i\omega)c_l^2 \beta} \left[\omega^2 - \left(\frac{c_l^2}{\gamma} + \frac{4\mu}{3\rho} \right) k_c^2 \right], \quad (11)$$

$$\Gamma_T = -\frac{\gamma}{(-i\omega)c_l^2 \beta} \left[\omega^2 - \left(\frac{c_l^2}{\gamma} + \frac{4\mu}{3\rho} \right) k_T^2 \right], \quad (12)$$

where $\gamma = C_p/C_v$ is the ratio of the specific heat capacities and β is the thermal expansivity. For liquids, the speed $c_l = c$ and the last term in the parenthesis is negligible. It should be noted that the thermal factors for liquids which were given in Refs. 10–13 were not exact, although very good approximations for liquids. The error occurs by the assumption that the biharmonic equation for the scalar wave potential is directly separable into thermal and compressional terms. This is not in fact the case. The correct thermal factors are given by Allegra and Hawley for solids, and these can be converted for liquids by dividing by the factor $(-i\omega)$, as given above.

From these expressions it can be seen that Γ_c is predominantly imaginary, and Γ_T is predominantly real. The functions b_1 – b_3 have both real and imaginary components. As a result, the most significant real and imaginary parts in the

numerator are already obtained in only the first term of the series. However, contributions of order $(k_c a)^2$ relative to unity were found to be significant numerically under conditions where $|k_s a|^2$ is small. The expanded numerator series appears in the term M in the solution below [Eq. (13)]. Similarly for the denominator; although the dominant real and imaginary parts were included in previous expansions, the later terms in the series in $k_c a$ make a significant contribution under some conditions, particularly, when the thermal and shear wavenumber-radius products are small. This was discovered numerically. Hence the series are again expanded to higher order terms in $k_c a$, leading to the expression Δ in Eq. (13). The new terms in the denominator are of order $(k_c a)^2$ compared with unity, through the ratio Γ_c/Γ_T . Hence, our new result for the thermal part of the coefficient is

$$A_{0T} = ik_c a \cdot \frac{\Gamma_c}{\Gamma_T} \cdot G_1 G_2 H \cdot \frac{M}{\Delta}, \quad (13)$$

where

$$H = \left[\frac{1}{(1 - k_T a i)} - \frac{\tan(k'_T a)}{\hat{\tau}[\tan(k'_T a) - k'_T a]} \right]^{-1}, \quad (14)$$

$$M = 1 - \frac{(k'_c a)^2}{6} \left[1 + \frac{2b_3}{(1 - k_T a i)G_1 \hat{\rho}} + \frac{8}{(k_s a)^2 G_2 \hat{\rho}} (\hat{\eta} - 1 + G_2 \hat{\eta}) \right], \quad (15)$$

$$\Delta = 1 - G_1 H \cdot \frac{\Gamma_c}{\Gamma_T} \left[1 - \frac{\hat{\Gamma}_c}{\hat{\rho}} \left(1 - \frac{4}{(k_s a)^2} \right) + \frac{b_3 \hat{\Gamma}_c}{(1 - k_T a i)G_1 \hat{\rho}} + ik_c a G_2 \right]. \quad (16)$$

The parameter b_3 is a function of the thermal properties, resulting from the stress components, defined by Pinfield¹⁵

$$b_3 = q_{T0} - \frac{1}{\hat{\tau} \hat{\Gamma}_T} \cdot \frac{k_T a}{k'_T a} \cdot \frac{h'_0(k_T a)}{h_0(k_T a)} \cdot \frac{j_0(k'_T a)}{j'_0(k'_T a)} \cdot q'_{T0}, \quad (17)$$

$$q_{T0} = 1 - 2 \frac{(k_T a)^2}{(k_s a)^2} [1 + h''_0(k_T a)/h_0(k_T a)], \quad (18)$$

$$q'_{T0} = \hat{\rho} - 2\hat{\rho} \frac{(k'_T a)^2}{(k'_s a)^2} [1 + j''_0(k'_T a)/j_0(k'_T a)], \quad (19)$$

where h_n, j_n are spherical Bessel and Hankel functions, respectively. Their ratios can be expressed analytically for the zero order functions, if required, so that

$$h''_0(k_T a)/h_0(k_T a) = -1 + \frac{2}{(k_T a)^2} - \frac{2i}{k_T a}, \quad (20)$$

$$j''_0(k'_T a)/j_0(k'_T a) = -1 + \frac{2}{(k'_T a)^2} - \frac{2}{k'_T a \tan(k'_T a)}. \quad (21)$$

However, the second of these equations leads to inaccurate results when calculating numerically, when $(k'_T a)$ is small, due to the small difference between the large second and third terms. In that region, $\tan(k'_T a)$ should be expanded for small arguments, so that the large canceling terms are removed analytically leaving the smaller remaining terms to obtain a more accurate numerical result.

The parameters G_1 and G_2 in Eqs. (13), (15), and (16) denote the fractional differences in thermal properties between the inner and outer media.

$$G_1 = 1 - \frac{1}{\hat{\tau} \hat{\Gamma}_T}, \quad G_2 = 1 - \frac{\hat{\Gamma}_c}{\hat{\rho}}. \quad (22)$$

The new result for the thermal term has been expressed here [Eq. (13)] in a way to demonstrate the correction of the previous results given most fully by Allegra and Hawley [Ref. 3 Eq. (13)], which in our notation is

$$A_{0T} = ik_c a \cdot \frac{\Gamma_c}{\Gamma_T} \cdot G_1 G_2 H. \quad (23)$$

Clearly, the new terms developed in the present work are incorporated in the parameters M and Δ of Eq. (13). However, the original analytical result, Eq. (23) was further approximated, both by the original workers [Ref. 2 Eq. (10.2) and Ref. 3 Eq. (19)] and by later workers, for example as quoted in Ref. 9 [Eq. (2)], giving

$$A_{0T} = i(k_c a)^3 \cdot \frac{(\gamma - 1)}{(k_T a)^2} \cdot \left(1 - \frac{\hat{\beta}}{\hat{\rho} \hat{C}_p} \right)^2 H. \quad (24)$$

To see how our new expression for the thermal part of the coefficient [Eq. (13)] relates to this previous result [Eq. (24)],⁹ the thermal components G_1 and G_2 are examined first. The thermal factors, Γ_c and Γ_T have the approximate form (for liquids)

$$\Gamma_c \approx \frac{k_c^2(\gamma - 1)}{\beta \sigma k_T^2}, \quad \Gamma_T \approx -\frac{1}{\beta \sigma}, \quad (25)$$

when $\left| \frac{k_c^2}{k_T^2} \right| \approx \frac{\omega \sigma}{v^2} \ll 1$ which is true over a very wide frequency range (up to $\sim 10^{11}$ Hz for water at 30 °C). Adopting these expressions, it can be shown that both G_1 and G_2 are approximately equal to the thermal property difference factor which appears in the previous analytical expression [Eq. (24)]. Thus

$$G_1 \approx G_2 \approx 1 - \frac{\hat{\beta}}{\hat{\rho} \hat{C}_p}. \quad (26)$$

Although this relationship is sufficiently accurate for the imaginary part of the scattering coefficient (relating to sound speed), it is not a sufficiently accurate approximation for the real part (relating to attenuation). Hence, the factors G_1 and G_2 should be retained as in Eq. (22) rather than approximated.

Further correspondence between the new and old expressions for the thermal part of the coefficient can be

seen by evaluating the ratio of the thermal factors for the compressional and thermal waves, which takes the approximate form

$$\frac{\Gamma_c}{\Gamma_T} = \frac{(k_c a)^2 (\gamma - 1)}{(k_T a)^2}. \quad (27)$$

In a series of numerical calculations, we found that even the approximate forms for G_1 and G_2 for the ratio of the thermal factors, do not give a sufficiently accurate result for the real part of the thermal coefficient, A_{0T} , to successfully calculate the attenuation. The new derivation for the explicit expression includes many more terms than those given previously, but calculations have shown that these can be significant under some conditions and must be included. Approximations which are valid for the imaginary part of the coefficient may not be sufficiently accurate for the real part of the coefficient. Many of the new terms include a factor of $(k_S a)^2$ which is imaginary. When compared with real terms, its magnitude may be small but it may be the dominant imaginary part of the expression. In addition, when the frequency is low or radius is small, terms in $(k_S a)^{-2}$ will become more significant and therefore must be included. Expressions which include the thermal wavenumber will also contribute both real and imaginary terms which may have been neglected previously on the basis of their absolute magnitude. The numerical calculations in Sec. IV demonstrate the conditions under which these terms make a significant contribution to the attenuation.

IV. NUMERICAL RESULTS

In order to demonstrate the improved accuracy of the new analytical coefficient and the errors in the previous expression, the ultrasound attenuation has been calculated numerically for a model emulsion of 20% v/v sunflower oil in water, with a range of particle sizes (monodisperse) and frequencies. Calculations were carried out using MATLAB® (R2009a) running under Windows XP and 64 bit precision, an accuracy of 2×10^{-16} . The attenuation was determined from the wavenumber of the emulsion, calculated using the Lloyd and Berry multiple scattering model.¹⁷ For the full ECAH calculations (using the boundary condition matrix inversion), coefficients up to order 2 were included, but for the calculations using the low-frequency analytical coefficients only the zero and first order coefficients were included. The analytical result for the A_1 coefficient is available in the original papers, and is given by McClements *et al.*⁹ No inaccuracies have been identified with this result, and an analytical development has yet to be carried out. Hence, this coefficient was used for the calculations from the “explicit” coefficients. The physical properties of the two components are given in Table I.^{6,19}

The results for the calculated attenuation are plotted against the particle radius, at different frequencies (Fig. 1). At each frequency, results are shown for the full ECAH solution (symbols), those using the old explicit solutions for A_0 and A_1 (dashed lines), and those using the new explicit solu-

TABLE I. Physical properties of the simulated sunflower oil in water emulsion components at 25°C. Sunflower oil properties were mostly determined experimentally,¹⁹ whereas data for water were obtained from property handbooks.⁶ f is frequency in Hz.

	Water	Sunflower oil
Ultrasound velocity (m s ⁻¹)	1497.0	1469.9
Density (kg m ⁻³)	997.0	920.6
Thermal expansivity (K ⁻¹)	0.00026	0.00071
Viscosity (Pa s)	0.00088	0.054
Specific heat capacity (J kg ⁻¹ K ⁻¹)	4178.2	1980
Thermal conductivity (W m ⁻¹ K ⁻¹)	0.5952	0.17
Attenuation factor (Np m ⁻¹)	$2.5 \times 10^{-14} f^2$	$2.76 \times 10^{-11} f^{1.7}$

tion for A_0 with the old explicit solution for A_1 (dotted lines). A very clear deviation between the full and explicit results is observed as the particle radius becomes larger, and this deviation occurs at a smaller radius as the frequency increases. This limit is a result of the breakdown of the long wavelength, or small $k_c a$ condition. The solid line cutting across the attenuation curves shows the condition $k_c a = 0.1$; all radii to the left of this and frequencies below it are at lower $k_c a$.

It would be expected that a low- $k_c a$ approximation to the coefficients would be valid over the entire range below this limit. However, a significant difference can be identified at the smallest radii between the attenuation calculated from the old explicit approximations for A_0 [Eqs. (10) and (24)] and the full ECAH solution. This inaccuracy was the reason for the development of new explicit solutions for the coefficient A_0 , which are presented in this paper. Calculations for model emulsions in which the oil had a lower viscosity than sunflower oil showed a much lower error in attenuation

FIG. 1. (Color online) Plot of attenuation against particle radius for a 20% monodisperse sunflower oil in water emulsion at various frequencies. Results are compared for (markers) the full ECAH model (taken to two orders); (dashed lines) the old analytical expressions for A_0 and A_1 ; (dotted lines) the new analytical expression for A_0 (still using the old expressions for A_1). The attenuation is calculated from the Lloyd and Berry scattering calculation to the second order in volume fraction. The frequencies are 1 MHz (circles), 5 MHz (crosses), 10 MHz (triangles), and 25 MHz (squares). The solid line with diamond markers cutting across the attenuation curves shows the condition $k_c a = 0.1$; all radii to the left of this and frequencies below it are at lower $k_c a$.

calculated from the old analytical expressions at smaller $k_c a$. Therefore, we conclude that it is a combination of small particle size and high dispersed phase viscosity, which contributed to the inaccuracy in the old analytical coefficients relating predominantly to the terms in $1/(k_s a)^2$ in Eqs. (9) and (13).

Results from these new coefficients [Eqs. (9) and (13)] are plotted as dashed lines and can be seen to coincide with the full ECAH solution over the whole radius and frequency range within the long wavelength region. Hence the inclusion of the additional terms in the coefficients has made a significant improvement in the approximate attenuation. It was noticeable during development that approximation of parameters, such as G_1 could not be used in the coefficient A_0 in order to achieve a sufficiently accurate calculation of the attenuation. This is largely because it is necessary to retain sufficient accuracy in both real and imaginary parts of each component term independently, rather than simply in the modulus of that component.

V. DISCUSSION AND CONCLUSIONS

New results have been presented for the analytical low-frequency zero-order scattering coefficient for a fluid sphere embedded in a fluid medium. These results are applicable in the long wavelength region and are derived by expressing the coefficient as a series in the wavenumber-radius product $k_c a$ and retaining only the first few terms. Similar analytical expressions which have been in use for many years have been found to be inaccurate under some conditions. By retaining some further terms in the expansions, which become significant when the radius is small, a new analytical solution for the coefficient has been derived [Eqs. (9) and (13) with Eq. (6)]. Numerical calculations of the attenuation in a model emulsion have demonstrated the accuracy of the new results when compared to the full ECAH model.

It may be argued that the differences between the new and old formulations are relatively slight, and the latter may be acceptable for “engineering” purposes such as the measurement of dispersed particle size distributions (PSDs). We have found that such measurements are surprisingly sensitive to errors in either measured or modeled attenuation and that this sensitivity justifies the new explicit approximation presented in this paper, particularly for very small particles (nano-emulsions) or where the larger particle sizes in poly-disperse emulsions are close to the condition $ka = 0.1$. Such

uncertainties in PSD measurement will form the basis of a new publication which is currently in preparation.

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