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# Ultrasound propagation in concentrated suspensions: shear-mediated contributions to multiple scattering

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**Abstract**

We report analytical and numerical results of a multiple scattering model applied to silica-in-water suspensions. We investigate the shear-mediated effects due to mode conversion between compressional and shear wave modes, not included in standard multiple scattering models. We identify the dominant scattering contributions and develop analytical forms for them. Numerical calculations demonstrate the contribution of the additional shear-mediated effects to the compressional wave speed and attenuation through the suspension. As concentration is increased, we incorporate third order terms in concentration to the expansion of the effective wavenumber of the compressional wave. The calculations are compared with previously published experimental data.

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*Keywords:* suspensions; multiple scattering; shear mediated effects; third order expansion; pair correlation function; low frequency

## 1. Introduction

Ultrasonic techniques offer many advantages for process monitoring suspensions of particles, as detailed for example by Challis et al. [1]. Their application depends on the accuracy of the models used to interpret the measured ultrasonic speed and attenuation spectra in terms of particle size, concentration and physical properties. Multiple scattering models such as Lloyd and Berry's [2] have been used with great success in relatively dilute suspensions (up to 10%w/w) for colloidal particles, but were found inadequate at higher concentrations, smaller particles, and

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low frequencies by Hipp et al. [3], and later as well by Challis and Pinfield [4]. The principal reason was believed by the last authors to be the neglected shear-mediated contributions to multiple scattering. The model presented by Luppé, Conoir and Norris [5] does take into account mode conversions at each scattering event, and we use it to investigate those shear-mediated effects to the compressional wave properties in concentrated suspensions of silica spheres in water on which experimental studies had been reported by Hipp et al. [3].

**Nomenclature**

| | |
|---|---|
| $a$ | radius of the silica spheres |
| $b$ | radius of exclusion ($b$=2$a$ in the numerical part) |
| $c$ | concentration of scatterers |
| $k_C$, $k_S$ | wavenumbers of the compressional, shear wave in the host medium in the absence of scatterers |
| $K_C$ | effective wavenumber of the coherent compressional wave |
| $T_n^{(pq)}$ | mode $n$ scattering coefficient of a single sphere; incident wave of type $p$, scattered wave of type $q$ |
| $G(0,n\mid 0,m\mid \nu)$ | Gaunt coefficient, as defined in Cruzan [6] |
| $j_n, h_n^{(1)}$ | spherical Bessel and Hankel functions of order $n$ |

## 2. The multiple scattering model for concentrated suspensions of silica spheres in water

The multiple scattering model is that described in Eqs.(29-32) in Luppé et al. [5], giving the low concentration asymptotic expansion of the compressional effective wavenumber $K_C$, up to order 2 in powers of $\varepsilon = -3ic/(\pi a^3)$, under the hole correction assumption that the mean density number of scatterers at some location $\vec{r}_2$ , provided one scatterer is known to be centered at $\vec{r}_1$ , is given by

$$n\left(\vec{r}_2 \mid \vec{r}_1\right) = \frac{3}{4}\frac{c}{\pi a^3}\left(1 + g\left(\left\|\vec{r}_2 - \vec{r}_1\right\|, c\right)\right) \quad , \tag{1}$$

with $g(r,c) = H(r-b)$, and $H$ the Heaviside function. As we are interested here in concentrated suspensions, we have pushed up to order 3 in concentration, following the procedure described in Norris and Conoir [7]:

$$\frac{K_C^2}{k_C^2} = 1 - 3i\frac{c}{k_C^3 a^3}\delta_1^{(CC)} - 9i\frac{c^2}{k_C^6 a^6}\left[\delta_2^{(CC)} + \delta_2^{(CS)}\right] - 27i\frac{c^3}{k_C^9 a^9}\left[\delta_3^{(CC)} + \delta_3^{(CS)}\right] + o\left(c^4\right) \quad . \tag{2}$$

with the first order term $\delta_1^{(CC)}$ and second order terms $\delta_2^{(CC)}$ and $\delta_2^{(CS)}$ obtained from Luppé et al. [5] formulas, and the third order terms by following the procedure described in Norris and Conoir [7].

The silica spheres have a 2$a$ = 300 nm diameter, and their physical properties, as well as those of water, are given in Challis et al. [1]. The density of silica varies with the degree of porosity, and the density was taken here as 2100 $kgm^{-3}$ at which the Lloyd and Berry model predictions agree with the experimental data of Hipp et al. [3] at the highest frequency and largest diameter shown in their paper (400 nm, 100 MHz). The attenuation of the coherent compressional wave is computed from Eq. (2) as a function of the concentration $c$ of spheres, for different frequencies, ranging from 2 MHz $\left(\left|k_C a\right| \approx 0.001\right)$ to 100 MHz $\left(\left|k_C a\right| \approx 0.06\right)$. The scattering coefficients are determined using the generalisation of the formulation of Epstein and Carhart [8] and Allegra and Hawley [9], as in Challis et al. [10], and the thermal waves in and outside the spheres are shown to be negligible. Analytical approximations are performed under the long compressional wavelength assumption, following the same procedure as in Pinfield [11], in order to retain only the dominant terms in Eq. (2).

The monopole compressional to compressional scattering coefficient is found negligible in comparison to its dipole counterpart, and the scattering coefficients that involve one shear wave at least dominate all others, so that Eq.(2) is approximated as

$$\frac{K_C^2}{k_C^2} \approx \left[\frac{K_C^2}{k_C^2}\right]_{LB} - 9i\frac{c^2}{k_C^6 a^6} T_1^{(CS)} T_1^{(SC)} \Delta_2\left(k_C, k_S, b\right) - 27i\frac{c^3}{k_C^9 a^9} T_1^{(CS)} T_1^{(SC)} T_1^{(SS)} \Delta_3\left(k_C, k_S, b\right) \quad ,(3)$$

where $\left[\frac{K_C^2}{k_C^2}\right]_{LB}$ is the Lloyd/Berry formulation up to second order in concentration, and including only partial waves of order 0 and 1; this involves transition factors $T_0^{(CC)}$ and $T_1^{(CC)}$. $\Delta_1$ and $\Delta_3$ are functions of $k_C$, $k_S$, and $b$.

## 3. Numerical study. Comparison with experiment

Figure 1 shows the attenuation curves obtained by Hipp et al. [3] with symbols, along with those obtained from Eq. (3), either truncated at order 2 in concentration (solid lines), or whole (dotted lines). Those obtained from the Lloyd and Berry model [2], which consists in truncating Eq.(2) at order 2 in concentration and neglecting the shear-mediated term $\delta_2^{(CS)}$ are drawn as well (dashed lines) for the sake of comparison.

Neglecting the shear-mediated effects as in Lloyd and Berry's model [2] (dashed lines in figure 1) provides quite a good estimation of the attenuation, as long as the concentration is lower than about 10 %. At higher concentrations, the experiment shows a quasi-parabolic dependence of the attenuation on the volume fraction, while the Lloyd and Berry model exhibits a quasi-linear variation that overestimates the attenuation at "low" frequency. Taking into account the shear - viscous wave in the second order term of the effective wavenumber expansion leads to better shaped curves, but with a too pronounced parabolic behaviour and underestimates the attenuation at concentrations larger than the maximum abscissa. The best agreement between theory and experiment is achieved by taking into account as well the third order terms in concentration, as in Eq. (3).

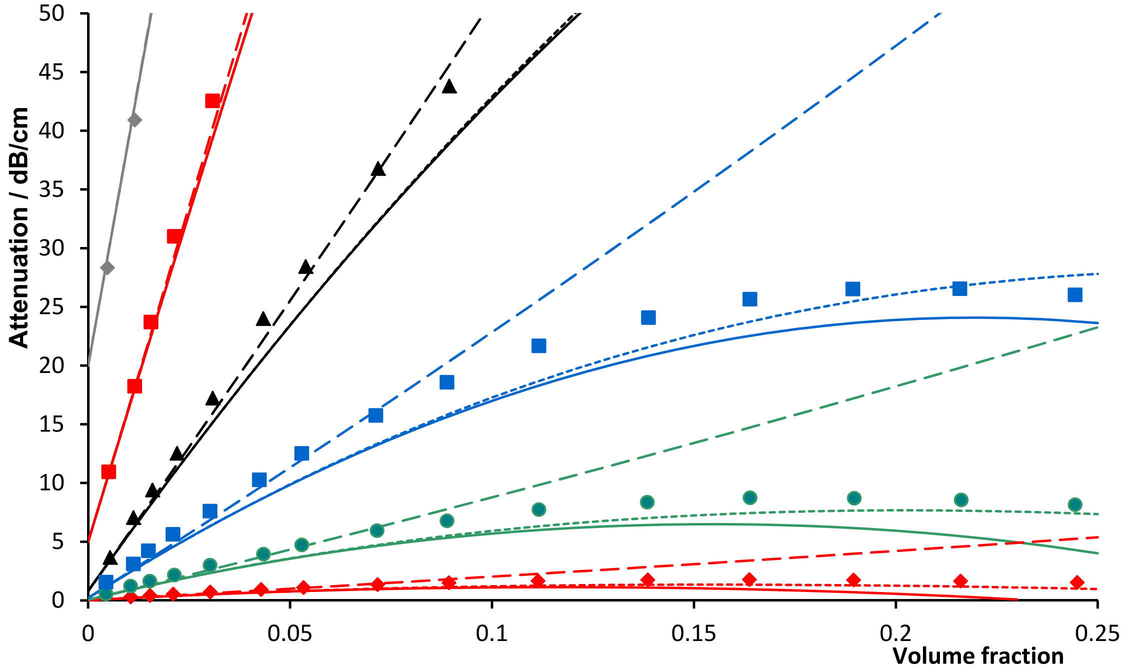


**Figure 1.** Attenuation as a function of concentration (volume fraction). Symbols : Hipp et al. [3] experiments. Solid lines : from Eq.(3), limited to second order in concentration. Dotted lines : from Eq.(3), up to third order in concentration. Dashed lines : Lloyd and Berry model [2]. The attenuation increases with frequency. Red: 2 MHz. Green : 5 MHz. Blue : 10 MHz. Black : 20 MHz. Red : 50 MHz. Gray : 100 MHz.

## 4. Conclusion

While much improved by accounting for both the shear wave effects and the third order terms in concentration, the model-estimated attenuation still lacks sufficient accuracy to be properly used in the monitoring of the suspension. For example, at 10 MHz, Eq. (3) predicts an attenuation around 2 dB/cm less than that measured by Hipp (10 % error)for a concentration equal to about 14 %. We believe this discrepancy between theory and experiment to be due to the fact that, as the concentration is increased, the hole correction becomes less and less reasonable, and a more realistic pair-correlation function should be taken into account. If, for example, the Virial series expansion given by Eq.(21) in Caleap et al. [12] is chosen, a new third order term in concentration appears, that, contrary to that of Eq. (3), involves the products of only two scattering coefficients, $T_1^{(CS)}$ and $T_1^{(SC)}$. First results show that its contribution to the wavenumber expansion might be of the same order of magnitude as $\delta_2^{(CS)}$ . That work is still in progress.

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