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Calculating the acoustic radiation force on spherical particles in a standing ultrasound wave field considering single and multiple scattering

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ABSTRACT

Ultrasound directed self-assembly (DSA) utilizes the acoustic radiation force (ARF) associated with a standing ultrasound wave to organize and orient particles dispersed in a fluid medium into specific patterns. The ARF is a superposition of the primary acoustic radiation force, which results from the incident standing ultrasound wave, and the acoustic interaction force, which originates from single and multiple scattering between neighboring particles. In contrast with most reports in the literature that neglect multiple scattering when calculating the ARF, we demonstrate that the deviation between considering single or multiple scattering may reach up to 100%, depending on the ultrasound DSA process parameters and material properties. We evaluate a theoretical case with three spherical particles in a viscous medium and derive operating maps that quantify the deviation between both scattering approaches as a function of the ultrasound DSA process parameters. Then, we study a realistic system with hundreds of particles dispersed in a viscous medium, and show that the deviation between the ARF resulting from single and multiple scattering increases with decreasing particle size and increasing medium viscosity, density ratio, compressibility ratio, and particle volume fraction. This work provides a quantitative basis for determining whether to consider single or multiple scattering in ultrasound DSA simulations.

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Ultrasound directed self-assembly (DSA) utilizes the acoustic radiation force (ARF) associated with a standing ultrasound wave to organize and orient particles dispersed in a fluid medium into specific patterns. The ARF is the superposition of the primary acoustic radiation force (PARF), which results from the incident standing ultrasound wave, and the acoustic interaction force (AIF), which originates from the acoustic interactions between neighboring particles. Researchers have studied the acoustic interactions between spherical particles using single scattering (see, e.g., Refs.26–31) and multiple scattering (see, e.g., Refs.32–36) and several publications argue that the effect of multiple scattering on the ARF is small and can be neglected in favor of single scattering.26,28,29,31 However, physically, it appears that multiple scattering becomes increasingly important with increasing particle volume fraction and local particle packing density, when particles are in close proximity and the acoustic interactions between them increase.37 Accurately calculating the ARF is essential when using ultrasound DSA in engineering applications. Consequently, several researchers have studied the effect of using different scattering approaches in ultrasound DSA simulations. Many research groups, including ours, have...
studied the effect of ultrasound DSA process parameters on the organization and orientation of particles in multiple dimensions in both inviscid and viscous media based on only the PARF, i.e., neglecting the AIF (e.g., spherical particles in 1D, 2D, and 3D; high aspect ratio particles in 2D and 3D). On the other hand, several researchers have also accounted for both the PARF and AIF using single scattering and multiple scattering. Specifically, Silva and Bruus theoretically demonstrated that the AIF between spherical particles in an inviscid medium subject to a plane ultrasound wave can be attractive or repulsive, accounting for single scattering only. Similarly, Lopes et al. analyzed the AIF between spherical particles in an inviscid medium subject to a plane ultrasound wave, but accounted for multiple scattering, and determined that its effect on the AIF depends on the relative position of the particles.

The literature does not provide any comparison between the ARF that acts on spherical particles in a viscous medium when accounting for single or multiple scattering. However, this information is important to accurately simulate the locations where particles organize during ultrasound DSA. Hence, the objective of this work is to quantify the relative contributions of single and multiple scattering in the calculation of the ARF for spherical particles in a viscous medium, as a function of the ultrasound DSA process parameters, including particle size, material properties, and medium viscosity.

We first consider a theoretical case with three spherical particles in a viscous medium and derive operating maps that quantify the deviation between the ARF based on single and multiple scattering as a function of the ultrasound DSA process parameters. Then, we consider a realistic system with hundreds of particles dispersed in a viscous medium relevant to, e.g., manufacturing engineered materials.

Figures 1(a)–(c) illustrate the theoretical model of a three-particle system to simulate the ARF for both single \( F_{\text{single}} \) and multiple scattering \( F_{\text{multiple}} \). Figure 1(a) shows a rectangular reservoir (gray) with two ultrasound transducers on opposing walls (orange) that establish a standing ultrasound wave within the viscous medium contained in the reservoir.

We simulate the incident velocity potential of a plane standing ultrasound wave as 
\[
\phi_{\text{inc}}(x) = e^{ik(x-L/2)} + e^{ik(x+L/2)},
\]
where \( L = 20\lambda \), \( \lambda = (c_0/c_m)/(1 - i\omega\tau_\text{visc})^{1/2} \) is the complex wave number that accounts for acoustic attenuation in the viscous medium with \( \tau_\text{visc} \) the viscous dissipation time to dampen the acoustic pressure to \( 1/e \) of its original value, \( c_m \) is the sound propagation velocity of the medium, \( \omega \) is the angular frequency, and \( k \) is the wavelength. The incident wave velocity 
\[
\mathbf{v}_{\text{inc}}(x) = \nabla \phi_{\text{inc}}(x).
\]

Figure 1(b) schematically illustrates single and multiple scattering between the particles. We calculate the ARF at the location of the first particle, which we refer to as the probe particle (maroon). We methodically position the second (orange) and third (gray) particles between a single node and an antinode of the standing ultrasound wave and calculate the ARF at the probe particle. Vectors \( \mathbf{r}_1 \), \( \mathbf{r}_2 \), and \( \mathbf{r}_3 \) describe the locations of the three particles with respect to the origin of the coordinate system in the simulation domain. Using single scattering, the incident wave scatters off the second and third particles once, and these scattered waves superimpose on the incident wave at the probe particle [teal arrows in Fig. 1(b)]. In contrast, considering multiple scattering, the incident wave scatters multiple times between the three particles, and multiple scattered waves superimpose on the incident wave at the probe particle [both teal and gray arrows in Fig. 1(b)].

We express the velocity potential of the ultrasound wave \( \phi_\text{inc} \) that scatters off the \( j \)-th particle located at \( \mathbf{r}_j \) and measured at the \( i \)-th particle located at \( \mathbf{r}_i \), accounting for both monopole [first term in Eq. (1)] and dipole [second term in Eq. (1)] scattering as follows.
\[ \phi_w(r, r_i) = \sigma \phi(r_i) G(r, r_i) + (Pv(r_i)) \frac{1}{k^2} \nabla G(r, r_i). \]  

(1)

Here, \( \phi(r_i) \) and \( v(r_i) = \nabla \phi(r_i) \) are the velocity potential and wave velocity at location \( r_i \), respectively, both of which derive from the Helmholtz equation that describes the ultrasound wave within the simulation domain. \( G(r, r_i) = \frac{e^{ik|r-r_i|}}{(4\pi|r-r_i|)} \) is the Green’s function for the Helmholtz equation in 3D, which relates the ultrasound wave at the particles and within the simulation domain. \( \nabla G(r, r_i) \) is the gradient of the Green’s function, and \( k = \Re(k) \) because viscous attenuation of the ultrasound wave is small \((\approx 0.08\%)\) over a short distance between a single node and antinode considered in this work. \( \sigma = 4\pi\mu k^2a^3/3 \) and \( P = -2\pi\mu k^2a^3 I_{3,3} \) are functions of the monopole \( f_1 \) and dipole \( f_2 \) scattering coefficients, respectively, where \( a \) is the particle radius, and \( I_{3,3} \) is a three-by-three identity matrix. The monopole scattering coefficient \( f_1 \) is a function of the compressibility potential \( \kappa \) and the dipole scattering coefficient \( f_2 \) is a function of the density ratio \( \rho_f/\rho_m \) and medium viscosity \( \eta_m \), where \( \beta \) and \( \rho \) are compressibility and density, and subscripts \( p \) and \( m \) refer to the particle and fluid medium. Figure 1(b) is a visualization of the magnitude of the monopole and dipole scattering velocity potentials \( \Re(\phi_1) \) for all particles (solid copper-color contour plots).

Thus, when considering multiple scattering, the total velocity potential \( \phi(r) \) at location \( r \) is the superposition of the incident velocity potential \( \phi_{inc}(r) \) and the sum of the velocity potentials \( \phi_w \) that result from the scattered ultrasound waves from all other particles, as well as at \( r_i \), i.e.,

\[ \phi(r) = \phi_{inc}(r) + \sum_{j \neq i} N \phi_w(r_i, r_j). \]  

(2)

\( N = 3 \) is the number of particles. Combining Eqs. (1) and (2) yields

\[ \phi(r_i) = \phi_{inc}(r_i) + \sum_{j \neq i} \left[ \sigma \phi(r_j) G(r, r_i) + (Pv(r_j)) \frac{1}{k^2} \nabla G(r, r_i) \right]. \]  

(3)

Furthermore, \( v(r_i) = \nabla \phi(r_i) \), i.e.,

\[ v(r_i) = v_{inc}(r_i) + \sum_{j \neq i} \left[ \sigma \phi(r_j) \nabla G(r, r_i) + \frac{1}{k^2} \nabla^2 G(r, r_i) Pv(r_j) \right]. \]  

(4)

where \( \nabla^2 G(r, r_i) \) is the Hessian of the Green’s function.\(^{55}\) We show the linear systems to calculate \( \phi(r) \) [Eq. (3)] and \( v(r) \) [Eq. (4)] in the supplementary material. The ARP at the location of the probe particle \( r \) is

\[ U(r) = \frac{4\pi}{3} a^3 \left( \frac{\rho_m}{2} \left( (ii\rho_m)^{op}(\phi(r_i))^3 \right) - f_2 \frac{3\rho_m}{4} \left( v_x(r_i)^3 \right) \right). \]  

(5)

where \( v_x(r_i) \) is the \( x \)-component of \( v(r_i) \) and \( \langle \cdot \rangle \) averages over a single wave period. The ARP with multiple scattering \( F_{multiple} = -\partial U(r_i)/\partial x \), using \( \phi(r_i) \) and \( v(r_i) \) from Eq. (5), and the ARP with single scattering \( F_{single} = -\partial U(r_i)/\partial x \), using \( \phi(r_i) \) and \( v(r_i) \) from Eq. (5). In addition, the PARF \( F_{inc} = -\partial U(r_i)/\partial x \), using \( \phi_{inc}(r_i) \) and \( v_{inc}(r_i) \) in Eq. (5), where \( v_{inc}(r_i) \) is the \( x \)-component of the incident velocity vector \( v_{inc}(r_i) \). Additionally, \( v_{inc}(r_i) = v_{inc}(r_i) = 0 \) for a plane standing ultrasound wave. The percent deviation between the ARF from single and multiple scattering, relative to the PARF is \( E = |F_{multiple} - F_{single}|/F_{inc} \). We validate our calculation of the ARF based on single and multiple scattering with the experimental results of Mohapatra et al.,\(^{30}\) who measured the ARF between pairs of spherical polystyrene particles in a standing ultrasound wave in water, based on the respective speeds with which they approach each other (see supplementary material).

Figure 1(c) illustrates the possible configurations of the probe, second, and third particles for which we evaluate \( E \). We fix the probe particle at \( x = 2/8 \), i.e., the middle between a node and antinode (white hollow marker), select ten locations for the second particle (black solid marker), and 100 \( \times \) 100 locations of the third particle (silver grid, see magnified inset image). We select the ten locations of the second particle based on symmetry. Five of these locations involve contact between the probe and the second particle, whereas the other five locations allow for the third particle to lie between the probe and the second particle. Figure 1(c) illustrates a typical result of \( E \) (solid color contour plot) for a single configuration of probe and second particles, and for 100 \( \times \) 100 locations of the third particle. The white area around the probe and second particles in Fig. 1(c) is the exclusion area that the third particle center cannot occupy to avoid overlap.

We determine the maximum deviation \( E_{max} = \max(E) \) as a function of ultrasound DSA process parameters and material properties, including the particle size \( a \), the medium viscosity \( \eta_m \), medium and particle density \( \rho_m \) and \( \rho_p \), and medium and particle compressibility \( \beta_m \) and \( \beta_p \). According to the Buckingham \( \pi \) theorem,\(^{32} \) four nondimensional parameters are required to describe this system: (i) the nondimensional particle size \( 0.05 < K_1 = ka < 0.20 \), which we select to satisfy the Rayleigh regime assumption \( (ka < 1) \), (ii) the nondimensional medium viscosity \( 0.00 < K_2 = \eta_m/\rho_m c_m \leq 0.27 \) that spans the viscosity range of different fluid media previously used with ultrasound DSA in the context of manufacturing engineered materials (e.g., water, photopolymer resin) \( 0 < \eta_m \leq 400 \text{ mPa s} \),\(^{37} \) (iii) the density ratio \( 0 \leq \rho_f/\rho_m \leq 2.5 \), and (iv) the compressibility ratio \( 0 < \beta_f/\beta_m \leq 2.5 \) that spans the density and compressibility ratios of relevant combinations of particles and media for manufacturing engineered materials.\(^{57} \)

Figures 1(a), 1(c), and 1(d) illustrate the theoretical model of a large-scale system to simulate the ARF accounting for both single \( F_{single} \) and multiple scattering \( F_{multiple} \), considering hundreds of randomly dispersed particles in a viscous medium. We illustrate the model with particle volume fraction \( \Phi = 5\% \) [Fig. 1(d)] and \( \Phi = 20\% \) [Fig. 1(e)]. We fix the probe particle at \( x = 2/8 \) (maroon particle) and randomly disperse \( N = 320 \) particles in the solution domain, which we scale to satisfy a specific particle volume fraction \( 5\% \leq \Phi \leq 20\% \). We compute the percent deviation \( E \) between the ARF for single and multiple scattering, and we measure the distance \( d \) between the probe particle and its closest neighbor. For each value of \( \Phi \), we perform 250 repeat simulations to quantify the variability of \( E \) as a result of the random positions of the particles that affects \( d \).

Figure 2 shows the ratio of the ARFs that account for scattering and the incident ultrasound wave, for (a) single \( F_{single}/F_{inc} \) and (b) multiple scattering \( F_{multiple}/F_{inc} \) in the three-particle system, and
tering is maximum in that configuration. Contact each other in the wave propagation direction because dipole scattering occurs when their line of centers is parallel to the wave propagation direction where $k_G r_1 > 1$. Furthermore, the angle between their lines of centers and the wave propagation direction affects the dipole scattering coefficient. Generally, maximum values of $F_{\text{single}} / F_{\text{inc}}, F_{\text{multiple}} / F_{\text{inc}},$ and $E_{\text{Max}}$ occur where particles contact each other in the wave propagation direction because dipole scattering is maximum in that configuration.

Figure 3 shows the maximum percent deviation $E_{\text{Max}}$ between the ARF based on single and multiple scattering, (a) as a function of nondimensional particle size $K_1$ and nondimensional medium viscosity $K_2$ for $\rho_p / \rho_m = 2.4$ and $\beta_p \beta_m = 0.029$, (b) as a function of density ratio $\rho_p / \rho_m$ and compressibility ratio $\beta_p / \beta_m$ for $K_1 = 1.2$, and $K_2 = 0.14$, where the particle is denser and less compressible than the fluid medium ($\rho_p / \rho_m \geq 1$ and $\beta_p / \beta_m \leq 1$), and (c) as a function of density ratio $\rho_p / \rho_m$ and compressibility ratio $\beta_p / \beta_m$ for $K_1 = 1.2$, and $K_2 = 0.14$, where the particle is less dense and more compressible than the fluid medium ($\rho_p / \rho_m \leq 1$ and $\beta_p / \beta_m \geq 1$).

We observe that $E_{\text{Max}}$ increases with decreasing $K_1$ and increasing $K_2$. Decreasing $K_1$ decreases the distance between neighboring particles, which increases the dipole scattering term in Eq. (1) that affects both single and multiple scattering and, thus, increases $E_{\text{Max}}$. Furthermore, $E_{\text{Max}}$ increases with increasing $\rho_p / \rho_m$ and increasing $\beta_p / \beta_m$. First, in the Rayleigh regime ($ka \ll 1$), dipole dominates monopole scattering because the gradient of the Green’s function $k^{-1} \nabla G(\mathbf{r}, \mathbf{r})$ in the x-direction is larger than the Green’s function $G(\mathbf{r}, \mathbf{r})$ itself [see Eq. (1)]. Second, maximum scattering between particles occurs when their line of centers is parallel to the wave propagation direction where $k^{-1} \nabla G(\mathbf{r}, \mathbf{r})$ is maximum. Under these conditions, dipole scattering becomes dominant.
circumstances, dipole scattering causes particles to repel, whereas monopole scattering causes particles to attract each other. Thus, monopole scattering and dipole scattering counteract each other in the $x$-direction, where the effect of dipole scattering is dominant. As a result, increasing $p_b/p_m$ and $\beta_p/\beta_m$ increases the dipole scattering coefficient $f_2$ and decreases the monopole scattering coefficient $f_1$. Since the scattering coefficients affect both single and multiple scattering, it increases $F_{\text{single}}/F_{\text{inc}}$ and $F_{\text{multiple}}/F_{\text{inc}}$ and, thus, $E_{\text{Max}}$. The results show that $E_{\text{max}}$ can reach up to 100%, which contrasts previous works in the literature concluding that multiple scattering is negligible compared to single scattering in the calculation of the AIF and ARF.\cite{dawson2024,oliver2018} However, these works focus on systems with two rather than three (or many) particles, which cannot entirely describe the effect of multiple scattering.

Figure 4(a) shows the percent deviation $E$ vs the shortest distance between neighboring particles $d$ for 250 different configurations of 320 randomly dispersed particles around the probe particle, for four different particle volume fractions $\Phi = 5\%$, 10\%, 15\%, and 20\%, and for $K_1 = 0.05$, $K_2 = 0.27$, $p_b/p_m = 2.4$, and $\beta_p/\beta_m = 0.029$. We use transparent solid data points so that increasing darkness signifies an increasing number of overlapping data points. The data shows that decreasing the distance $d$ between the probe particle and its nearest neighbor increases the deviation $E$, independent of $\Phi$, because the distance between particles is one of the main parameters that drives the relative importance between monopole and dipole scattering (Fig. 2). Additionally, Fig. 4(b) shows the probability density function of $E$, and its mean value $E_{\text{mean}}$ for 250 configurations of 320 randomly dispersed particles around the probe particle, for the results of Fig. 4(a). The likelihood of a small distance $d \approx 2a$ increases with increasing $\Phi$ because it increases the number of non-overlapping particles in the solution domain. Thus, multiple scattering becomes increasingly important with increasing $\Phi$ because the interactions between particles increase.

This methodical study highlights the importance of considering multiple scattering effects in ultrasound DSA simulations under certain conditions; the operating maps of Fig. 3 guide the choice between both scattering approaches in ultrasound DSA simulations. However, limitations still exist. We simulate the incident wave as a perfect plane standing wave, but realistic transducers cannot generate a perfect wave, and reflections from the near-field interfere with the reservoir walls and potentially affect the results. Furthermore, the medium viscosity may induce nonlinear phenomena such as streaming at the reservoir walls and microstreaming around the particles, which could affect the ARF.\cite{dawson2024,oliver2018} In practice, imperfect dispersion, shape, and size of the spherical particles as well as thermal fluctuations in the viscous medium also affect the ARF.

We conclude that the deviation $E$ between the ARF derived from single and multiple scattering may reach up to 100\%, depending on the ultrasound DSA process parameters and material properties. Thus, neglecting multiple scattering in favor of computationally efficient single scattering is not always correct. Physically, the distance between spherical particles, the angle between their lines of centers relative to the wave propagation direction, the ultrasound DSA process parameters (nondimensional particle size $K_1$, nondimensional viscosity $K_2$, particle volume fraction $\Phi$), and the material properties (density ratio $p_b/p_m$ and compressibility ratio $\beta_p/\beta_m$) determine the relative magnitude of monopole and dipole scattering between spherical particles in a standing ultrasound wave and, thus, $E$. The maximum deviation $E_{\text{max}}$ occurs when particles contact each other in the wave propagation direction because this configuration causes the largest dipole scattering between particles. Furthermore, $E$ increases with decreasing $K_1$ and increasing $K_2$, and with $p_b/p_m$ and increasing ratio $\beta_p/\beta_m$ largely driven by increasing dipole scattering that affects both single and multiple scattering and, thus, increases $E$. Comparing the fundamental three-particle system to a large-scale system shows that increasing $\Phi$ increases $E$ because it increases the number of particles in a constant
control volume, decreases the distance between them, and increases both single and multiple scattering.

See the supplementary material shows the derivation of Eqs. (3) and (4) and illustrates experimental validation of the model.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Soheyl Noparast: Conceptualization (equal); Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (equal); Visualization (lead); Writing – original draft (lead); Writing – review & editing (equal). Fernando Guevara Vasquez: Conceptualization (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Supervision (supporting); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). Mathieu Francoeur: Conceptualization (equal); Funding acquisition (equal); Visualization (supporting); Writing – review & editing (supporting). Bart Raeymaekers: Conceptualization (lead); Formal analysis (equal); Funding acquisition (equal); Methodology (equal); Project administration (lead); Supervision (lead); Visualization (supporting); Writing – original draft (equal); Writing – review & editing (lead).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.


