Supplementary Material for

Particle Dynamics in Sheared Particulate Suspensions

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Appendix A

In this appendix, we explain the method used for relating the components of the average particle velocity and its variance to the values measured experimentally using Dynamic Sound Scattering in a cylindrical cell with a focusing transducer.

(1) Principles of the method to obtain average $\vec{q}$

In this appendix, we derive an expression for the average scattering wave vector $\langle \vec{q} \rangle$ when an ultrasonic wave from a spherically focusing transducer is incident on a cylindrical cell. The method is based on a ray approach, since this simplified approach leads naturally to a description in terms of scattering angles for the experimental geometry. We are interested in the possible values of $\vec{q} = \vec{k} - \vec{k}'$, where $\vec{k}$ is the incident wave vector on a scatterer and $\vec{k}'$ is the scattered vector. Only some $\left(\vec{k}, \vec{k}'\right)$ combinations are relevant due to the finite extent of the transducer. We found all such $\vec{k}$, $\vec{k}'$ by integrating over the surface of the transducer. For each point $\left(\xi, \zeta\right)$ on the transducer surface, two wave vectors $\vec{k}_L$ and $\vec{k}_S$ are possible results of the ray propagating from $\left(\xi, \zeta\right)$ in the direction of the focus into the interior of the sample, passing through the cell wall in either longitudinal (*L*) or shear (*S*) mode.

Geometrically it is clear that the scattered vectors $\vec{k}'_L$ and $\vec{k}'_S$ that arrive back at the transducer are anti-parallel to the set of $\vec{k}_L$ and $\vec{k}_S$:

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$$\vec{k}'_L(\xi,\zeta) = -\vec{k}_L(\xi,\zeta); \qquad \vec{k}'_S(\xi,\zeta) = -\vec{k}_S(\xi,\zeta).$$

The various transmission and reflection coefficients bringing a given ray from (ξ,ζ) into the sample can be combined to give transfer functions $T_L(\xi,\zeta)$ and $T_S(\xi,\zeta)$ that express the relative contribution of the given $\vec{k}$ inside the sample. Outgoing transfer functions $T'_L(\xi,\zeta)$ and $T'_S(\xi,\zeta)$ express the proportion of scattered waves that arrive at the transducer as a function of (ξ,ζ) (*i.e.*, as a function of $\vec{k}'$).

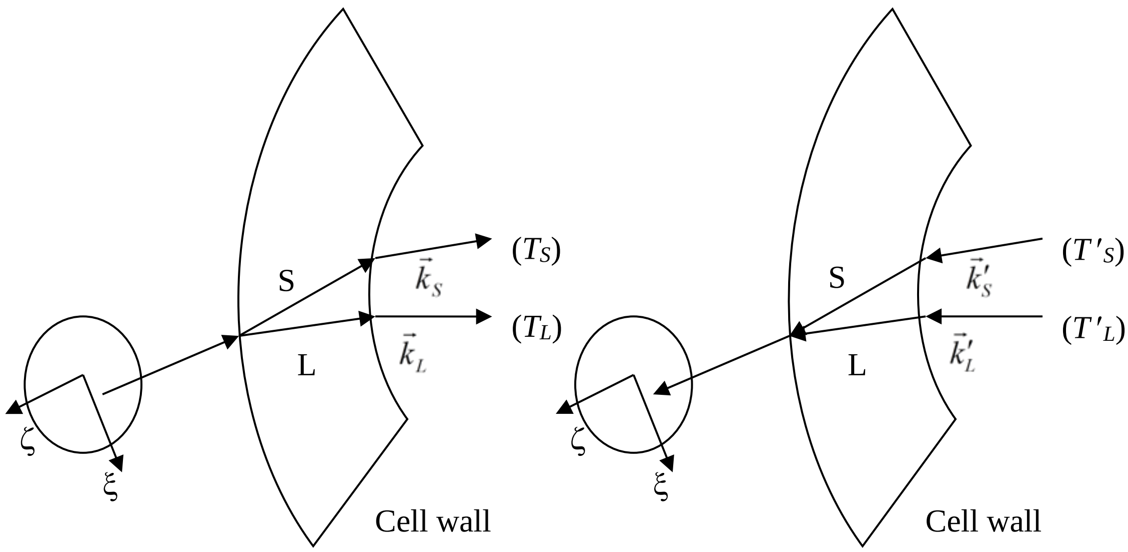

Figure A1. Ray diagram illustrating refraction at the cell walls

For scattering particles moving with $\vec{V}$ we measure an average of $(\vec{q}\cdot\vec{V})^2 = q_r^2 V_r^2 + q_\phi^2 V_\phi^2 + q_z^2 V_z^2$ (simplification possible since $\overline{V}_r = \overline{V}_z = 0$) and wish to know the relative contributions of V_r^2, V_ϕ^2 and V_z^2; thus, we are interested in the average or net q_i^2 (for $i = r$, ϕ, z) that results from the range of $\vec{k}$, $\vec{k}'$ produced and observed by the transducer. Hence the required quantity is

$$\text{net } q_i^2 \equiv Q_i = \sum_{\text{modes}} \frac{1}{A^2} \int_S d\zeta\, d\xi\, T(\zeta,\xi) \int_S d\zeta' d\xi'\; T'(\zeta',\xi') \left(k_i(\xi,\zeta) - k'_i(\xi',\zeta')\right)^2,$$

where the integrals are over the surface S of the transducer $(\xi^2 + \zeta^2 < R)$ and the sum indicates that we must account for L and S modes in both directions. For computation, we have:

$$\begin{aligned} Q_i = \frac{1}{A^2} \sum_{\text{modes}} \Bigg[&\left(\int_S d\zeta\, d\xi T(\zeta,\xi) k_i^2(\xi,\zeta) \right) \left(\int_S d\zeta' d\xi' T'(\zeta',\xi') \right) \\ &+ \left(\int_S d\zeta\, d\xi T(\zeta,\xi) \right) \left(\int_S d\zeta' d\xi' T'(\zeta',\xi') k_i'^2(\xi',\zeta') \right) \\ &-2 \left(\int_S d\zeta\, d\xi\, T(\zeta,\xi) k_i(\xi,\zeta) \right) \left(\int_S d\zeta' d\xi' T'(\zeta',\xi') k'_i(\xi',\zeta') \right) \end{aligned}$$

Since $k'(\xi,\zeta) = -k(\xi,\zeta)$

$$Q_i = \frac{1}{A^2}\sum_{\text{modes}}\left[\left(\int_S d\zeta d\xi\, T\, k_i^2\right)\left(\int_S d\zeta d\xi\, T'\right) + \left(\int_S d\zeta d\xi\, T\right)\left(\int_S d\zeta d\xi\, T'\, k_i^2\right)\right.$$
$$\left. +2\left(\int_S d\zeta d\xi\, T\, k_i\right)\left(\int_S d\zeta d\xi\, T'\, k_i\right)\right]$$

Accounting for all pairs of modes leaves us with six integrals:

$$Q_i = \frac{1}{A^2}\left[I_i^1 I_i^{3\prime} + I_i^3 I_i^{1\prime} + 2 I_i^2 I_i^{2\prime}\right]$$

where

$$I_i^1 = \int_S d\zeta d\xi \left(T_L k_{i,L}^2 + T_S k_{i,S}^2\right)$$
$$I_i^2 = \int_S d\zeta d\xi \left(T_L k_{i,L} + T_S k_{i,S}\right)$$
$$I_i^3 = \int_S d\zeta d\xi \left(T_L + T_S\right)$$

and similarly for $I_i^{j\prime}$, replacing *T* with *T′*.

In the computations, six sums are kept, calculated with

$$S_i = \sum_{\xi,\zeta}\begin{bmatrix} T_L & T_S \\ T_L' & T_S' \end{bmatrix}\begin{bmatrix} 1 & k_{i,L} & k_{i,L}^2 \\ 1 & k_{i,S} & k_{i,S}^2 \end{bmatrix} \approx \frac{m}{A}\begin{bmatrix} I_i^3 & I_i^2 & I_i^3 \\ I_i^3{}' & I_i^2{}' & I_i^3{}' \end{bmatrix},$$

where *m* is number of elements in the summation.

After summing, the Q_i are formed from the S matrix, e.g.

$$Q_i = \frac{1}{m^2}\left[S(1,1)S(2,3) + S(2,1)S(1,3) + 2S(1,2)S(2,2)\right].$$

A similar calculation yields the net values of q_i:

$$P_i \equiv \text{net } q_i = \frac{1}{A^2}\sum_{\text{modes}}\left(\int_S d\xi d\zeta\ T k_i \int_S d\xi d\zeta\ T' + \int_S d\xi d\zeta\ T \int_S d\xi d\zeta\ T' k_i\right)$$
$$= \frac{1}{A^2}\left(I_i^2 I_i^3{}' + I_i^3 I_i^2{}'\right) = \frac{1}{m^2}\left[S(1,2)S(2,1) + S(1,1)S(2,2)\right].$$

A computer program was written in MATLAB to calculate Q_i and P_i. The output of the program is

$$\begin{bmatrix} Q_r & Q_\phi & Q_z & P_r & P_\phi & P_z \end{bmatrix}.$$

This result can be used to find the variance or average velocity along the r, ϕ and z directions from measurements along 3 different transducer orientations, by solving the matrix equation involving the Q's or P's. For example for the variance, we have to solve

$$\begin{bmatrix} Q_r^1 & Q_\phi^1 & Q_z^1 \\ Q_r^2 & Q_\phi^2 & Q_z^2 \\ Q_r^3 & Q_\phi^3 & Q_z^3 \end{bmatrix} \begin{bmatrix} (\delta V_r)^2 \\ (\delta V_\phi)^2 \\ (\delta V_z)^2 \end{bmatrix} = \begin{bmatrix} q^2 \left\langle \delta V^2 \right\rangle^1 \\ q^2 \left\langle \delta V^2 \right\rangle^2 \\ q^2 \left\langle \delta V^2 \right\rangle^3 \end{bmatrix}$$

[calculated for 3 geometries (1,2,3)] [Desired (common)] [Measured for 3 geometries (1,2,3)]

In the next sections, the various steps that need to be considered in the calculation of the transfer functions are described.

(2) Conversion from the experimental angle describing transducer orientation (*a*) to the incident angle used for calculations (*m*)

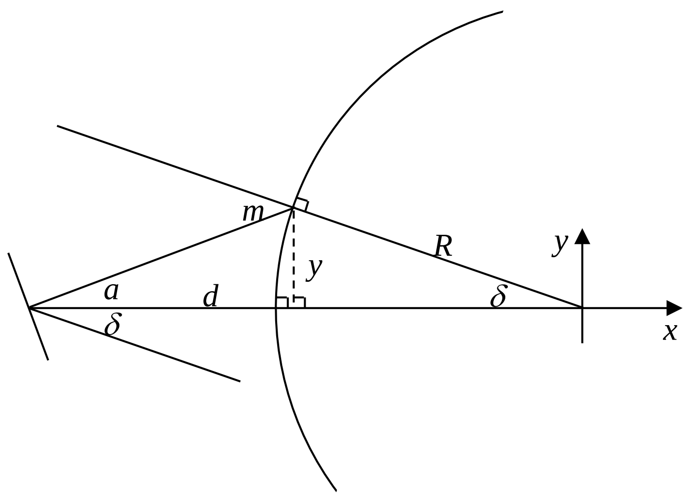

Figure A2. Finding the incident angle in terms of the transducer angle.

The geometry relating these angles (a, m) is defined in Figure A2.
To relate the angles a to m, we use the x and y coordinates, and intersect the circle $x^2 + y^2 = R$ with a line having slope $\tan a \equiv T_a$, passing through $\left(-\left(R+d\right),0\right)$, *i.e.*,

$$y = T_a\left(x + \left(R+d\right)\right)$$
$$x^2 + T_a^2\left(x^2 + \left(R+d\right)^2 + 2x\left(R+d\right)\right) = R^2$$

$$x^2\left(1+T_a^2\right)+x\left(2\left(R+d\right)\right)+\left(R+d\right)^2 T_a^2-R^2=0$$

The next step is to solve for *y*, so

$$y/T_a-\left(R+d\right)=x$$

$$\left(\frac{y}{T_a}\right)^2-\left(R+d\right)^2-\frac{2y\left(R+d\right)}{T_a}=R^2-y^2$$

$$y\left(1+T_a^2\right)-2yT_a\left(R+d\right)+\left(\left(R+d\right)^2-R^2\right)T_a^2=0$$

$$y=\frac{2T_a\left(R+d\right)\pm\sqrt{4T_a^2\left(R+d\right)^2-4\left(1+T_a^2\right)\left(d^2+2RD\right)T_a^2}}{2\left(1+T_a^2\right)}$$

The solution for *y* that we are seeking is the closer of the two possibilities to 0, *i.e.*, the appropriate root has the negative sign for $T_a > 0$ and the positive sign for $T_a < 0$. We can generalize this by factoring out the T_a^2 from the radical:

$$y=\frac{T_a\left(R+d\right)-T_a\sqrt{\left(R+d\right)^2-\left(1+T_a^2\right)\left(d^2+2Rd\right)}}{\left(1+T_a^2\right)}$$

or

$$\frac{y}{R}=\frac{T_a}{1+T_a^2}\left[1+\frac{d}{R}-\sqrt{1-2\left(\frac{d}{R}\right)T_a^2-\left(\frac{d}{R}\right)^2 T_a^2}\right]$$

Now $\sin\delta = y/R$ and $m=a+\delta$, so $m=a+\sin^{-1}\left(y/R\right)$.

(3) Ray from transducer to cell wall.

The transducer was divided into small elements and the calculation was done for each element position on the transducer surface.

Let us create the ray from this element of the transducer to the (0,0,0) point on the surface of the cell.

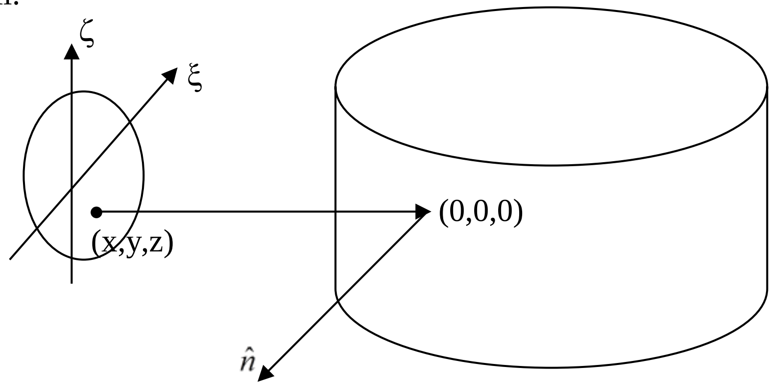

Figure A3. Diagram illustrating the ray from an element on the transducer to the point (0,0,0).

This ray is described by Euler angles

$$a = \tan^{-1}(x/z)$$
$$b = \tan^{-1}(y/z)$$

Refraction at the interface is treated with plane wave equations. The angle of incidence θ is given by

$$\tan\theta = \sqrt{\tan^2 a + \tan^2 b}$$

And, upon entering the solid, there are both shear and longitudinal modes produced.

The Euler angles are obtained from the refraction angles using the fact that the incident and refracted rays lie in a plane with the normal to the surface, and thus, *e.g.*,

$$\frac{\tan a}{\tan b} = \frac{\tan a_L}{\tan b_L}$$

So the refraction routine determines:

$$\tan\theta = \sqrt{\tan^2 a + \tan^2 b}$$
$$\sin\theta = \frac{\tan\theta}{\sqrt{1+\tan^2\theta}}$$
$$\sin\theta_L = \frac{n_1}{n_{2,L}}\sin\theta \quad ,$$

where θ_L is angle of refraction.

Euler angles are efficiently found considering:

$$\tan a = x/z$$
$$\tan a_L = x_L/z_L$$
$$\tan\theta = \frac{\sqrt{x^2+y^2}}{z}$$
$$\tan\theta_L = \frac{\sqrt{x_L^2+y_L^2}}{z_L}$$

and $x/y = x_L/y_L$ so

$$\frac{\tan\theta_L}{\tan\theta} = \frac{\sqrt{x_L^2\left(1+(y_L/x_L)^2\right)}}{\sqrt{x^2\left(1+(y/x)^2\right)}}\frac{z}{z_L} = \frac{x_L}{z_L}\frac{z}{x} = \frac{\tan a_L}{\tan a}$$

i.e., $\tan a_L = \tan a\left(\frac{\tan\theta_L}{\tan\theta}\right)$

and similarly $\tan b_L = \tan b\left(\frac{\tan\theta_L}{\tan\theta}\right)$.

(4) Transmission coefficient for solid *S* and *L* modes

We calculate θ_L and θ_S via the usual refraction conditions, e.g., $n_{2,L}\sin\theta_L = n_1 \sin\theta$.

The impedances are $z = \dfrac{\rho}{n\cos\theta}$.

Then

$$
\begin{aligned}
t_L &= \frac{2z_L \cos(2\theta_s)}{z_L\cos^2(2\theta_s) + z_s\sin^2(2\theta_s) + z_1}\frac{\rho_1}{\rho_2}\\
t_S &= \frac{-2z_S \sin(2\theta_s)}{z_L\cos^2(2\theta_s) + z_s\sin^2(2\theta_s) + z_1}\frac{\rho_1}{\rho_2}\\
r &= \frac{1-2z}{z_L\cos^2(2\theta_s) + z_s\sin^2(2\theta_s) + z_1}.
\end{aligned}
$$

These are the field transmission coefficients; the normalization condition is

$$
t_L^2\left(\frac{n_{2,L}\rho_2\cos\theta_{2,L}}{n_1\rho_1\cos\theta}\right) + t_S^2\left(\frac{n_{2,S}\rho_2\cos\theta_{2,S}}{n_1\rho_1\cos\theta}\right) + r^2 = 1
$$

(These modified coefficients are referred to as “intensity” transmission and refraction coefficients.)

We are also interested in the transmission of “outgoing” rays – those originating in the interior. We examine the longitudinal and shear waves in the cell wall that would yield a refracted angle θ in the water, in which the transducer is located, surrounding the cell.

(5) “Deviation of the normal” for the 2nd interface

We now have *S* and *L* rays in the cell wall. To refract at the next interface, we must account for the curvature of the cell walls. This is referred to as the “deviation of the normal”; the question is: by what angle do the normals to the cell walls differ (due to the curvature) for a ray that has traveled obliquely through the wall with initial angle of refraction α (see Figure A4)?

The curvature will only affect the α Euler angle ($\beta' = \beta$). This is the same problem as the conversion from “*a*” to “*m*” coordinates:

$$
\cos\delta' = \frac{T_\alpha}{1+T_\alpha^2}\left[1 + \frac{d}{R} - \sqrt{1 - 2\left(\frac{d}{R}\right)T_\alpha^2 - \left(\frac{d}{R}\right)^2 T_\alpha^2}\right]
$$

with $T_\alpha = \tan\alpha$.

Then $\alpha' = \alpha + \delta'$. Note δ' is different for the *S* and *L* waves: $\alpha_L' = \alpha_L + \delta_L'$, $\alpha_S' = \alpha_S + \delta_S'$. (See Figure A4 for the definitions of these angles.)

Refraction at the cell-interior interface is once again calculated using refraction. Note that since $\delta_L' \neq \delta_S'$, the refraction angles inside the cell will not be equal in general, $\alpha_L'' \neq \alpha_S''$, (they are probably pretty close though).

Once again the transmission coefficients are found for shear→liquid, longitudinal→liquid for "ingoing" rays, and liquid→shear, liquid→longitudinal for "outgoing" rays. The final ingoing and outgoing, shear and longitudinal transmission coefficients, are found by:

$$T_{in,L} = T_{in,L}^{1\to 2} \times T_{in,L}^{2\to 3}$$

$$T_{out,L} = T_{out,L}^{2\to 1} \times T_{out,L}^{3\to 2}$$

etc.

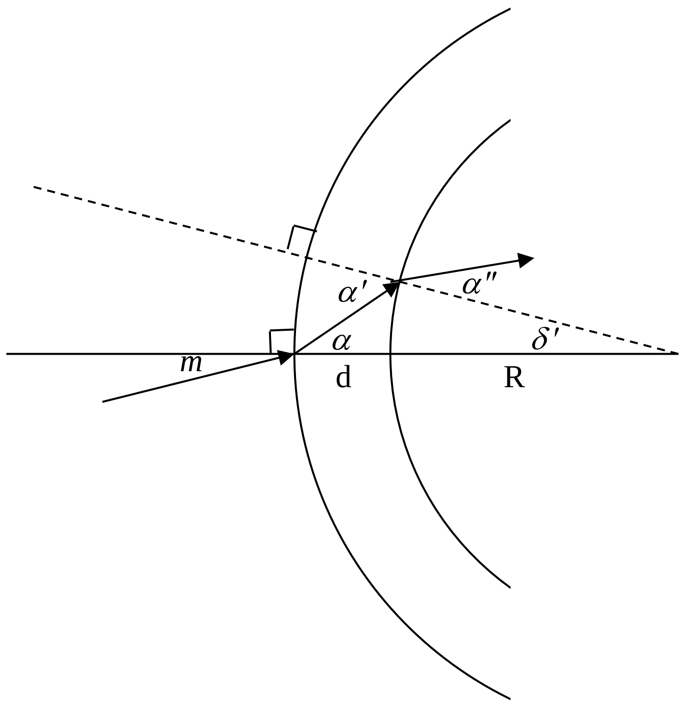

Figure A4: Refraction angles for transmission through the cell wall.

(5) $\vec{k}$ inside the cell

We have delineated the propagation of rays to the interior of the sample, and we need the $\vec{k}$ components here. These are different for *L* and *S* waves.

Normalized so $|\vec{x}| = 1$, the x'', y'', z'' components (see Figure A5) are found from the Euler angles

$$x_L'' = \frac{\tan \alpha_L''}{\sqrt{\tan^2 \alpha_L'' + \tan^2 \beta_L'' + 1}}$$

$$y_L'' = \frac{\tan \beta_L''}{\sqrt{\tan^2 \alpha_L'' + \tan^2 \beta_L'' + 1}}$$

$$z_L{}'' = \frac{1}{\sqrt{\tan^2 \alpha_L + \tan^2 \beta_L + 1}}$$

where the angles are defined in Figure A5.

The correspondence between the (x'', y'', z'') coordinates and the cell (r, ϕ, z) coordinates is

$$(\hat{r}, \hat{\phi}, \hat{z}) \sim (\hat{z}'', \hat{x}'', \hat{y}'').$$

We have thus determined the $\vec{k}$ vector (or the direction of such) of ray propagation from (ξ, ζ) in the transducer to the focus inside the cell via each mode in solid. This is related to the $\vec{k}$ vector of the ray at the focus which would propagate to (ξ, ζ) on the transducer by, *e.g.*, $\vec{k}_{L,out} = -\vec{k}_{L,in}$. For ($\xi$, ζ) we have thus found $\vec{k}_{in}$ and $\vec{k}_{out}$ for L and S modes, as well as the transmission coefficients, ingoing and outgoing, for both modes.

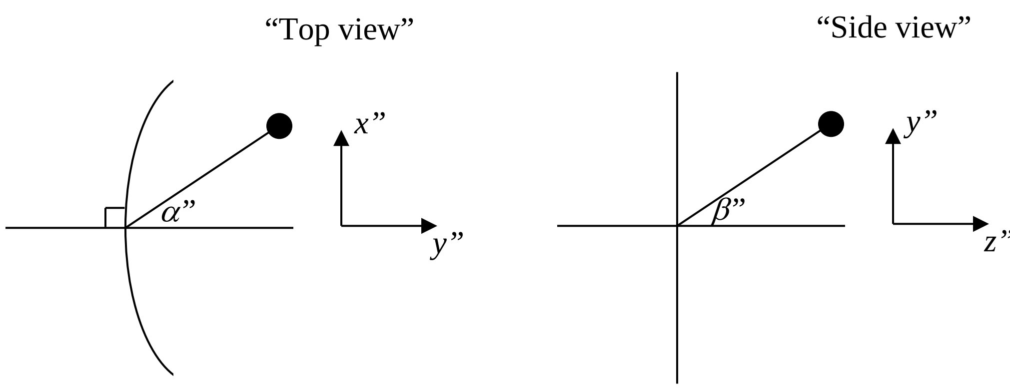

Figure A5: Refraction angles and double primed coordinate system for the interior of the cell.