Effective magnetic susceptibility of suspensions

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We characterize how suspensions of magnetic particles in a liquid respond to a magnetic field in terms of the effective magnetic susceptibility \( \chi_{\text{eff}} \) using inductance measurements. We test a model that predicts how \( \chi_{\text{eff}} \) varies due to demagnetization, as a function of sample aspect ratio, particle packing fraction, and particle aspect ratio \( [1] \). For spherical particles or cylindrical particles aligned with external magnetic field, the model can be fitted to the measured data with agreement within 17%. However, we find that the random alignment of particles relative to the magnetic field plays a role, reducing \( \chi_{\text{eff}} \) by a factor of 3 in some cases, which is not accounted for in models yet.

While suspensions are predicted to have \( \chi_{\text{eff}} \) that approach the particle material susceptibility in the limit of large particle aspect ratio, instead we find a much smaller particle aspect ratio where \( \chi_{\text{eff}} \) is maximized. A prediction that \( \chi_{\text{eff}} \) approaches the bulk material susceptibility in the limit of the packing fraction of the liquid-solid transition also fails. We find \( \chi_{\text{eff}} \) no larger than about 4 for suspensions of iron particles.

I. INTRODUCTION

Suspensions of magnetic particles in a liquid can be controlled by an applied magnetic field, a property that is taken advantage of for example in the fields of ferrohydrodynamics [2] and magnetorheology [3]. The parameter that directly controls the force applied by a magnetic field in these cases is the effective magnetic susceptibility \( \chi_{\text{eff}} \). If the suspensions are also conducting, magnetohydrodynamic effects can occur, such that a magnetic field can in principle be generated by the conducting fluid flow, and the magnetic field can deflect the conducting flow via a Lorentz force, effects whose magnitude scales with \( 1 + \chi_{\text{eff}} \) [1, 2]. While these phenomena are not easily achieved with known fluids – where pure conducting liquids generally have a magnetic susceptibility \( \chi \ll 1 \) – there is potential that if a material can be designed with large enough \( \chi_{\text{eff}} \geq 1 \), these phenomena could be more easily observed on a laboratory or device scale of order 10 cm [3].

Our goal is to determine how the effective magnetic susceptibility \( \chi_{\text{eff}} \) depends on the particle properties of suspensions. In particular, we would like to obtain larger values of \( \chi_{\text{eff}} \) to make such suspensions useful for producing magnetohydrodynamic phenomena on the laboratory scale.

The effective susceptibility \( \chi_{\text{eff}} \) is defined by the proportionality \( \chi_{\text{eff}} = \phi M/H_{\text{app}} \), where \( H_{\text{app}} \) is an externally applied magnetic field, \( M \) is the magnetization per unit volume of magnetic material, and \( \phi \) is the volume fraction of the magnetic particles. Note that the factor of \( \phi \) in the expression differs from traditional definitions in pure materials where susceptibility is defined per unit volume of magnetic material, as a pure material is 100% magnetic material. Instead we define \( \chi_{\text{eff}} \) as susceptibility per unit volume of sample, since we are interested in the force from an applied magnetic field on the sample as a whole. For linear magnetic materials \( \chi_{\text{eff}} \) is independent of \( H_{\text{app}} \), in practice this tends to be the case for small \( H_{\text{app}} \) before the magnetization begins to saturate.

Locally, the magnetic susceptibility \( \chi = M/H \) is considered a bulk material property depending on the local magnetic field \( H \). In contrast, \( \chi_{\text{eff}} \) as a macroscopic parameter can be much smaller than \( \chi \) due to demagnetization, an effect in which the induced magnetic dipole creates an additional magnetic field \( DM \) (where \( D \) is called the demagnetization factor) that opposes \( H_{\text{app}} \). The net magnetic field inside the material \( H_{\text{app}} - DM \) that determines the net local magnetization \( M \) is less than \( H_{\text{app}} \), resulting in \( \chi_{\text{eff}} \) being smaller than \( \chi \). The effective susceptibility can then be written such that the demagnetization is a correction factor on the material susceptibility:

\[
\frac{\phi}{\chi_{\text{eff}}} = \frac{1}{\chi} + D. \tag{1}
\]

It is well-known for single-piece solid magnets, for example, that \( D \) depends on the shape of the magnet, in particular \( D \) is small in the limit of long, thin magnets aligned with the applied magnetic field, (in this limit \( \chi_{\text{eff}} \) approaches the material susceptibility \( \chi \)). For single-piece solids, unless the aspect ratio of the material is

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extremely large, $\chi_{\text{eff}} \ll \chi$ and to a good approximation $\chi_{\text{eff}} \approx \phi / D$. $\chi_{\text{eff}}$ has been calculated for many particle shapes. For example, for a spherical particle $D = 1/3$, resulting in a maximum $\chi_{\text{eff}} \approx 3$ as long as $\chi \gg 3$. For such geometries with aspect ratio close to 1, the demagnetization effect can be considered a dominating factor determining $\chi_{\text{eff}}$, rather than a small perturbation on the material susceptibility $\chi$.

Demagnetization factors are less well-understood for systems of random arrangements of particles such as suspensions. With many particles, the demagnetization factor $D$ can depend on geometries of both the particles and the sample as a whole, as well as positions and alignments of particles relative to each other and the applied magnetic field.

For example, in the magnetorheological effect, a suspension exposed to an applied magnetic field develops a yield stress. The magnitude of this yield stress scales roughly as the force of the induced dipole-dipole interaction between the particles in suspension, which is proportional to $\chi^2_{\text{eff}}$ [9]. It has been observed that large-aspect ratio rod-shaped particles exhibit a larger yield stress than spherical particles, which was qualitatively attributed to the demagnetization effect [9]. However, this is not quantitatively understood, due to the lack of a model that relates this yield stress to $\chi_{\text{eff}}$ and the demagnetization effect.

For randomly packed spherical particles, it has been theoretically argued that the demagnetization factor is $D = 1/3 + \phi (D_g - 1/3)$, where $D_g$ is the global demagnetization factor based on the geometry of the sample and $\phi$ is the volumetric packing fraction [10]. $D_g$ was assumed to be the same as the demagnetization factor for a single particle of the same shape. A numerical calculation confirmed this model is a good approximation within 3% for a sample of randomly packed spherical particles for sample aspect ratios $\gamma_g = 0.5$ to 1, and packing fractions $\phi$ from 0.4 to 0.6 [11]. It remains to be seen how well the prediction holds over a wider parameter range, in particular at larger $\gamma_g$ where $\chi_{\text{eff}}$ is expected to be larger.

A more general model takes advantage of the fact that exact expressions can be found for homogeneously magnetized ellipsoids of revolution to obtain an expression for ellipsoidal particles homogeneously dispersed in any non-magnetic medium (including suspensions) in which particles are aligned with each other and the external magnetic field [12]. This demagnetization factor is

$$D = D_p (1 - \phi) + D_g \phi$$

where $D_p$ is the demagnetization factor of each particle (they are assumed to be identical). $D_p$ may be different from the global demagnetization factor $D_g$, and so is an unknown function of particle geometry. An expression for $\chi_{\text{eff}}$ can be obtained by combining Eq. (1) and (2) which simplifies if demagnetization effects are as significant as they are for typical for single-piece ferromagnetic materials in the limit where $\chi \gg \chi_{\text{eff}}$ to

$$\chi_{\text{eff}} \approx \frac{\phi}{D_p (1 - \phi) + D_g \phi}.$$  

To our knowledge, it has not yet been tested whether this model captures the effects of different particle shapes on $\chi_{\text{eff}}$ — specifically there is no model or data on how $D_p$ depends on particle aspect ratio or other parameters.

An alternate model designed for the limit of high packing fraction $\phi$ assumes that magnetic field lines tend to go from one ferromagnetic particle to another along regions of high susceptibility, and thus concentrate their density in paths along the shortest distances between particles [12]. It predicts that $\chi_{\text{eff}}$ diverges as the gaps between magnetic particles go to zero, approaching the material susceptibility $\chi$, according to

$$\chi_{\text{eff}} = \frac{1}{1 - (\phi / \phi_c)^{1/3}} - 1,$$

while $\chi_{\text{eff}} \ll \chi$. The critical packing fraction $\phi_c$ physically corresponds to the liquid-solid transition where particles with no long-range repulsions just barely touch. While this model has been tested at low $\phi$, it has not been tested at $\phi$ within 0.08 of the liquid-solid transition where the divergence would be expected to produce $\chi_{\text{eff}} \gg 1$ [12, 13], so it is not yet known if this divergence can be realized in suspensions.

In this manuscript, we test the above predictions for $\chi_{\text{eff}}$ for suspensions of cylindrical and spherical particles in cylindrical samples, by measuring $\chi_{\text{eff}}$ over a wide range of packing fraction $\phi$ up to $\phi_c$, sample aspect ratio $\gamma_g$, and particle aspect ratio $\gamma_p$. The remainder of the manuscript is organized as follows. We first describe the suspensions used in Sec. II A We describe the gravimeter we built to measure $\chi_{\text{eff}}$ in Sec. II B and its calibrations in Secs. II C and II D. We test the linearity of the magnetic response of the suspensions in current and frequency in Sec. III A. Measurements of $\chi_{\text{eff}}$ as a function of $\phi$, $\gamma_g$ and $\gamma_p$ are reported in Sec. III B. We use this to fit the demagnetization functions $D_p$ and $D_g$ from Eq. as a function of aspect ratios $\gamma_p$ and $\gamma_g$, respectively, in Sec. III C. Finally in Sec. III D we vary particle aspect ratio $\gamma_p$ in suspensions of randomly arranged particles to test whether particle alignment with the magnetic field plays an important role in $\chi_{\text{eff}}$, an effect which was not accounted for in Eq. 2.

II. MATERIALS AND METHODS

A. Materials

We suspended iron particles (density 7.834 kg/m$^3$ and purity 99.5%) of mean diameter 29 $\mu$m, where 90% of particle diameters are within the range 18-40 $\mu$m, purchased from Chemicalstore.com. The particles are nearly
FIG. 1. (a) The circuit diagram of the gradiometer used to measure the effective susceptibility $\chi_{eff}$ of a sample based on the change in mutual inductance of a solenoid pair. (b) A diagram (not to scale) that shows a cross-section of the coils and sample.

spherical with a standard deviation of 4% in the diameter. We use these nearly spherical particles in experiments unless otherwise specified. The suspending liquid was a eutectic alloy of gallium and indium known as eGaIn, which was produced as described in [7]. We used a liquid metal for its potential in magnetohydrodynamic applications. The properties of the liquid metal are not expected to be important here, other than the effect of its conductivity $3.40 \times 10^6 \text{ S/m}$ [14] contributing to stronger eddy currents that could reduce $\chi_{eff}$ at high frequencies of applied alternating magnetic field (see Sec. III.A). Samples were kept in an acid bath to prevent oxidation of the metals [3]. Dry granular samples were obtained by mixing the iron particles with non-magnetic sand.

In either case, the packing fraction $\phi$ was obtained by measuring masses of the constituent materials, and using density to convert to a packing fraction by volume of the magnetic material (iron) divided by the total volume taken up by the sample. In the case of dry granular samples, the total volume of the sample was measured directly as the volume taken up in the sample container, which includes some air.

B. Experimental setup

Measurements were taken using a gradiometer setup which consists of two pairs of inductor coils shown in Fig. [1]. The gradiometer measures $\chi_{eff}$ of a material sample based on how it changes the mutual inductance between two surrounding coils $P_1$ and $S_1$. The sample sits in a cylindrical tube which is placed inside the secondary coil $S_1$, while coil $S_1$ is inside the primary coil $P_1$. There is another nominally identical set of coils $P_2$ and $S_2$. An alternating current $I_p$ is applied at frequency $f$ (angular frequency $\omega = 2\pi f$) through the primary coils $P_1$ and $P_2$, while the induced voltage $\epsilon_{ind}$ is measured across both the secondary coils $S_1$ and $S_2$. The secondary coils are linked in the opposite direction in the circuit such that the mutual inductances of each pair of primary and secondary coils – $M_1$ and $M_2$, respectively – cancel in their contribution to the measured $\epsilon_{ind}$ when there is no sample inside coil $S_1$. In ideal theory, the induced voltage is then proportional to $\chi_{eff}$. In practice, the two pairs of coils are not identical which we account for with a small correction factor $\Delta M = M_1 - M_2$. Furthermore, there is a background voltage noise $\epsilon_{noise}$ measured when there is no sample and no applied current. The theory of induction allows derivation of an expression for the induced voltage $\epsilon_{ind}$

$$\epsilon_{ind}^2 = \epsilon_{noise}^2 + \omega^2 I_p^2 [\alpha M_1 \chi_{eff} + \Delta M]^2 . \quad (5)$$

where $\alpha$ is the fraction of volume of coil $S_1$ filled by the sample. This expression assumes that the background noise is distributed among all phases, which differs from the fixed phase of the induction signal, so that the root-mean-square values of the respective contributions to the induced voltage are added in quadrature.

The geometric parameters of the system are as follows. The primary coil $P_1$ has length $L_p = 127.0 \pm 0.2$ mm, diameter $d_p = 50.8 \pm 0.2$ mm, and $N_p = 332 \pm 22$ turns of wire. The secondary coil $S_1$ has length $L_s = 25.4 \pm 0.2$ mm, diameter $d_s = 14.8 \pm 0.2$ mm, and $N_s = 190 \pm 14$ turns of wire. The coils $S_2$ and $P_2$ are nominally identical to $S_1$ and $P_1$, respectively. All samples were prepared in cylindrical containers of length $L$ that satisfies $L_s < L < L_p$ so that they were fully contained in the uniform region of the applied field and their edge effects have a minimal effect on the flux seen by coil $S_1$. The filling fraction of the coil $S_1$ is then given by $\alpha = d^2 / d_s^2$, where $d$ is the diameter of the sample. We aligned the sample vertically within coil $S_1$ by finding the position of maximum measured induced voltage, as misalignment along the axis of the cylinder results in a reduced signal. The samples had inner diameter $d = 10.2 \pm 0.1$ mm unless otherwise specified, for aspect ratio corresponding to a typical filling factor $\alpha = d^2 / d_s^2 = 0.471$. The sample aspect ratio is given by $\gamma_p = L / d$.

Here we summarize some typical electrical measurement parameter values and errors. We report root-mean-square values for all of our measurements of both alternating current and voltage throughout the paper. For our measurements, the applied alternating current is typically $I_p = 65 \pm 0.5$ mA (corresponding to an 0.8% error) unless otherwise noted, where the error is given by the manufacturer (Agilent model 34401A multimeter). We typically report measurements at frequencies $f$ ranging from 200 to 2000 Hz, and $\chi_{eff}$ is calculated from Eq. 5 using an unweighted average over this frequency range unless otherwise specified. At these typical measurement values and when $\chi_{eff} \geq 1.2$, for example, we measure $\epsilon_{ind} \geq 23$ mV for different samples, with an uncertainty $\leq 0.2\%$ ($\geq 0.05$ mV) based on the 0.06%$\epsilon_{ind} + 0.04$ mV reported by the manufacturer, which is generally less than the uncertainty on the current measurement. The noise term $\epsilon_{noise}$ is due to electronic noise, and as such, varies when the measurement equipment is on. It is thus
measured as $\epsilon_{\text{ind}}$ at a weak signal with frequency $f = 5$ Hz at $I_p = 65$ mA. We measured $\epsilon_{\text{noise}} = 3$ mV on average, with a standard deviation of 0.4 mV over the course of a series of experiments shown in one plot, or 1 mV over the longer time scale of different measurement series. When added in quadrature as in Eq. 5, this leads to an error on $\epsilon_{\text{ind}}$ of less than 0.2% for $\chi_{\text{eff}} \geq 1.2$ for example, which is small compared to the other errors for these typical measurement parameters. This error becomes dominant when the signal is smaller, notably where we test the linearity of the signal at small values of $I_p$ or $f$ in Sec. III A or small $\phi$ where $\chi_{\text{eff}} \ll 1$. Similarly, the absolute error on $\chi_{\text{eff}}$ from the error of $\Delta M$ is 0.01, or equivalently less than 0.8% of $\alpha M_1 \chi_{\text{eff}}$ when $\chi_{\text{eff}} \geq 1.2$ for the typical measurement parameters (see Sec. III C on how values of $M_1$ and $\Delta M$ are obtained). Thus, the largest systematic source of error in calculating $\chi_{\text{eff}}$ from Eq. 5 unless otherwise noted typically comes from the 0.8% on the applied current $I_p$ for our typical parameters and $\chi_{\text{eff}} \geq 1.2$.

When we repeated measurements by turning off the electronics, taking a sample container out from inside the coils, putting the sample back, and turn on the electronics again, the run-to-run standard deviation was 2.5% for suspensions and granular samples, and 0.2% for macroscopic solid pieces. The larger run-to-run variation of suspensions and powders may come from the rearrangement of particles as the sample containers are disturbed, but it is smaller than the 6% standard deviation observed in numerical simulations [11].

C. Inductance calibration

To provide calibration values of $M_1$ and $\Delta M$ in Eq. 5, we measure the mutual inductances of each coil independently, in each case removing the other coil from the circuit and measuring without a sample. In these cases, the measured voltage is expected to be

$$\epsilon_{\text{ind}}^2 = \epsilon_{\text{noise}}^2 + \omega^2 I_p^2 M_i^2,$$

where $i = 1$ or 2 is the coil pair index number. Measurements of $\epsilon_{\text{ind}}^2$ are shown as a function of $\omega^2 I_p^2$ in Fig. 2 for both coil pairs. We fit a linear function plus a constant to each to obtain the slopes $M_1$ and $M_2$, respectively. The error bars in the figure represent the sum of a 0.2% standard deviation of multiple repetitions and a 0.8% systematic error. To obtain a fit with a reduced Chi-squared of 1 (where the reduced Chi-squared value of a fit corresponds to the mean-square difference between the data and fit, normalized by the error), we adjust the percentage input errors to 1.6% and 1.9% for coil pairs 1 and 2, respectively. The fit yields $M_1 = (1.245 \pm 0.004) \times 10^{-4}$ H and $M_2 = (1.253 \pm 0.004) \times 10^{-4}$ H. These measured values are consistent with the expected theoretical value $M = N_p N_s A_s / L_p = (1.2 \pm 0.1) \times 10^{-4}$ H based on the dimensions of the setup, where $A_s = \pi d_s^2 / 4$ is the cross-sectional area of the secondary coil. The difference between these measured mutual inductances is $\Delta M = M_2 - M_1 = (8 \pm 6) \times 10^{-7}$ H. These values $M_1$ and $\Delta M$ are used as calibrations to calculate $\chi_{\text{eff}}$ from Eq. 5.

D. Susceptibility calibration

We used single-piece solid cylindrical samples to calibrate $\chi_{\text{eff}}$ measurements in our setup. To account for the demagnetization effect, we use for reference a numerical simulation of the demagnetization factor $D$ for single-piece cylindrical samples of various aspect ratios $\gamma$.
To calibrate our setup, we measured \( \chi_{\text{eff}} \) of single-piece solid samples as a function of aspect ratio \( \gamma \). Solid circles: material susceptibility \( \chi = 850 \). Open square: \( \chi = 24500 \). Open diamond: \( \chi = 2000 \). Lines: fits of numerical simulation results from Chen et al. [15], for the different material susceptibilities \( \chi \) as given in the legend to obtain reference curves \( \chi_{\text{ref}} \). 

From Chen et al. [15], shown in Fig. 3 We fit the function

\[
D = A \gamma^n.
\]

To this data, over the range 0.7 < \( \gamma \) < 50, which covers our measurement range. We adjusted the input error to be 8% to obtain a reduced \( \chi \)-squared of 1, yielding \( A = 0.31 \pm 0.01 \) and \( n = -1.12 \pm 0.02 \).

To calibrate our setup, we measured \( \chi_{\text{eff}} \) of single-piece solid samples with different dimensions (the specific lengths and diameters are indicated in Fig. 4) and materials. Measured \( \chi_{\text{eff}} \) are shown in Fig. 4 at different aspect ratios \( \gamma \) for ferrite (\( \chi = 850 \), Fair-Rite Products Corp.), mu-metal (\( \chi = 2000 \), Aperam), and Permalloy (\( \chi = 24500 \), National Magnetics Group, Inc.). The plotted errors are the sum of the 0.2% run-to-run variation and 0.8% systematic error. A reference curve \( \chi_{\text{ref}} \) is shown in Fig. 4 for each material, which is calculated by inserting the fit function for \( D \) (Eq. 7) into Eq. 1 with \( \gamma = 1 \). Since these \( \gamma \) are all much greater than 1, the predicted \( \chi_{\text{ref}} \) curves are all close to each other. For aspect ratio \( \gamma \geq 10 \), the measured \( \chi_{\text{eff}} \) values collapse onto the reference curves \( \chi_{\text{ref}} \) within a root-mean-square difference of 7%. However, for \( \gamma \leq 5 \) the measured \( \chi_{\text{eff}} \) are about 30% smaller than the reference curve.

To come up with an appropriate calibration adjustment, we first consider that sample aspect ratio may not be the primary parameter which it could depend on. In the ideal theory assumed in Eq. 5 if \( L_s \ll L \ll L_p \), the magnetic field inside \( S_1 \) is expected to be uniform. In practice, fringe effects may add a correction. To come up with a calibration adjustment as a function of the sample length \( L \), we replot our measurements of \( \chi_{\text{eff}} \) for single-piece samples from Fig. 4 normalized by the reference curve \( \chi_{\text{ref}} \) as a function of the sample length \( L \) in Fig. 5. Different sample diameters \( d \) are indicated in the figure legend with uncertainties of 0.2 mm. A systematic dependence on \( L \) is observed in Fig. 5 similar to the trend in Fig. 4. In contrast, there is no systematic trend in \( d \), as some points for each value of \( d \) are in each of the lower and upper ranges of \( \chi_{\text{eff}}/\chi_{\text{ref}} \). This confirms the calibration should be made as a function of \( L \), but not as a function of \( d \). For \( L \geq 54 \) mm, the reference curve agrees with our measurement within a root-mean-square difference of 7% (a 7% error bar is plotted in Fig. 5 to see this). However, for \( L \leq 42 \) mm (= 1.7\( L_s \)), \( \chi_{\text{ref}} \) is an average of 40% ± 4% larger than \( \chi_{\text{eff}} \). Based on these results, we introduce a calibration factor in which the following measurements for \( \chi_{\text{eff}} \) are shifted upwards by a factor of 1.4 for samples with \( L \leq 42 \) mm. We note that most of our samples in later measurements have \( L \geq 54 \) mm, and this calibration factor only needs to be applied to a few of our shortest samples, specifically for aspect ratio \( \gamma_g = 2.5 \) in Fig. 8 samples with \( \gamma_g < 5 \) in Fig. 9 and samples with aspect ratio \( \gamma_g = 4.1 \) in Figs. 13 and 14. Based on the variation of 7% around the reference curve observed here, we also introduce an error of 7% from unknown sources when comparing samples in all following measurements of \( \chi_{\text{eff}} \).

### III. RESULTS

#### A. Linearity of magnetic response

To test the linearity of the magnetic properties of the materials with frequency, some examples of the measured susceptibility \( \chi_{\text{eff}} \) are shown for dry granular materials
FIG. 6. Examples of $\chi_{\text{eff}}$ as a function of frequency $f$. Solid symbols: suspensions of iron particles in eGaIn at $\phi = 18\%$ (circles) and $\phi = 40\%$ (squares). Open symbols: dry granular materials at $\phi = 20\%$ (up-pointing triangles) and $\phi = 43\%$ (down-pointing triangles). $\chi_{\text{eff}}$ reaches a plateau for $f < 2000\,\text{Hz}$. The vertical lines indicate the bounds of the frequency range where $\chi_{\text{eff}}$ is averaged over for measurements reported in other plots.

and suspensions as a function of frequency $f$ in Fig. 6 at sample aspect ratio $\gamma_g = 11$, length $L = 112.2\,\text{mm}$, and packing fractions $\phi$ shown in the legend. The error bars plotted the quadratic sum of the $2.5\%$ run-to-run standard deviation and the $0.4\,\text{mV}$ error on the noise voltage measurements, the latter of which tends to lead to a large error at low frequencies where the signal is weak. A plateau in $\chi_{\text{eff}}$ is found at frequencies $f < 2000\,\text{Hz}$ for all suspensions of nearly spherical particles reported in this paper. At higher frequencies, $\chi_{\text{eff}}$ decreases, qualitatively similar to the frequency response of other magnetic materials. The decrease starts at lower frequencies for suspensions than dry granular materials, which may be expected due to stronger eddy currents in the higher conductivity suspensions. At frequencies $f < 200\,\text{Hz}$, the data remain consistent with the plateau, however there are large relative uncertainties in this range due to the low voltage signal. Thus, in other plots in this paper, we report the averaged $\chi_{\text{eff}}$ over the range of $200\,\text{Hz}$ to $2000\,\text{Hz}$ as the representative value for the low-frequency plateau, unless we specify otherwise that we found the low-frequency plateau in a different range. This could introduce an error if there is a trend in $\chi_{\text{eff}}$ with frequency, as seen for $\phi = 18\%$ suspension in Fig. 6. In this case, which is comparable to the worst case, using the mean of $\chi_{\text{eff}}$ for frequencies in the range of $200\,\text{Hz}$ to $2000\,\text{Hz}$ can underestimate a fit in the zero-frequency limit by up to $3\%$, which is negligible compared to the $7\%$ error we use when comparing samples.

We next test whether the magnetic response is linear in the applied magnetic field $H_{\text{app}}$ (equivalently, whether $\chi_{\text{eff}}$ is independent of $H_{\text{app}}$), and whether the suspensions behave more like paramagnetic or ferromagnetic materials. We plot the magnetization per unit volume of sample $\phi M = \chi_{\text{eff}} H_{\text{app}}$ as a function of applied magnetic field $H_{\text{app}}$. Closed circles: increasing $H_{\text{app}}$ (or $I_p$). Open triangles: decreasing $H_{\text{app}}$ (or $I_p$). Line: linear fit. The suspension behaves as a linear paramagnetic material, with no hysteresis or significant remnant magnetization.

FIG. 7. Magnetization per unit volume of sample $\phi M = \chi_{\text{eff}} H_{\text{app}}$ of suspensions as a function of applied magnetic field $H_{\text{app}}$. Closed circles: increasing $H_{\text{app}}$ (or $I_p$). Open triangles: decreasing $H_{\text{app}}$ (or $I_p$). Line: linear fit. The suspensions are known to behave more like paramagnetic or ferromagnetic materials. We plot the magnetization per unit volume of sample $\phi M = \chi_{\text{eff}} H_{\text{app}}$ vs. the applied magnetic field $H_{\text{app}} = I_p N_p / L_p$ in Fig. 7 for a suspension with $\phi = 0.34$, $\gamma_g = 2.5$, and $L = 25.40\,\text{mm}$. We performed these measurements with histories of both increasing and decreasing applied current $I_p$ ($\propto H_{\text{app}}$). It is seen in Fig. 7 that these ramps give equivalent results, indicating a lack of hysteresis in the measured range. To test the linear response, we fit a linear function with a constant offset to these data where the random error is the quadratic sum of the $2.5\%$ run-to-run standard deviation and the $0.4\,\text{mV}$ random error on voltage measurements, which yields a reduced Chi-squared of $0.8$. The consistency of the linear fit with the data confirms the data are consistent with a $\chi_{\text{eff}}$ independent of $H_{\text{app}}$ over this range, verifying the linearity assumed in deriving Eq. 6. The error bars plotted in Fig. 7 include both these systematic and random errors. The constant offset in the linear fit was $5\,\text{A/m}$, which is consistent with $M = 0$ at $H = 0$ within the error of $12\,\text{A/m}$ on $M$ at that point due mainly to the $0.5\,\text{mA}$ systematic error on the current, so there is no resolvable remnant magnetization. These properties suggest the suspensions behave as linear paramagnetic materials in this range, which is simpler for both modeling and control, despite the fact that the particles themselves are ferromagnetic. The suspensions are known behave like a ferrofluid such that after an applied magnetic field is removed, the particles separate and flow like a liquid. This allows the ferromagnetic particles to move around in the liquid and reorient more freely than magnetic domains in a solid to avoid hysteresis and remnant magnetization.
exhibit a slightly smaller $\chi_{\text{eff}}$ than dry granular materials. Lines: fit of Eq. 8 for $\gamma_g = 11$ (solid line) and $\gamma_g = 2.5$ (dashed line), where fit parameters are obtained from simultaneously fitting data of Figs. 8, 9, and 10. Eq. 4 is shown for $d = 3.7 \pm 0.1$ mm for $\gamma_g > 22$. Measurements for $\gamma_g < 5$ have been adjusted upward by 40% according to the calibration in Fig. 4. At small $\gamma_g$, $\chi_{\text{eff}}$ increases with $\gamma_g$, and reaches a plateau for $\gamma_g \gtrsim 10$. For comparison to previous work in Fig. 3, we show numerical simulation results of randomly packed spherical particles at $\phi = 40\%$ [11] (Bjork et al. reported demagnetization factors $D$ [11], which we converted to $\chi_{\text{eff}} = \phi/D$. Their aspect ratio was defined as the inverse of our aspect ratio definition.). The simulation data follow the same trend as ours.

To test the dependence of $\chi_{\text{eff}}$ on particle aspect ratio $\gamma_p$ in Eq. 3 which assumes that particles are aligned with the applied magnetic field, we made dry samples of stacked cylindrical particles where the particles were forced to be aligned with the applied magnetic field. To make such aligned samples while holding $\gamma_g$ and $\phi$ constant, we cut a 130 mm long cylindrical ferrite rod into collections of gradually smaller pieces to obtain a series of decreasing $\gamma_p$. Each piece was nearly cylindrical, with roughness on a scale of 1 mm at the two ends of the cylinder due to the cutting process. The packing fraction $\phi$ ranged from 100% to 97% due to some loss of material. This resulted in a number of pieces ranging from 1 for the largest $\gamma_p$ to 32 for the smallest $\gamma_p$ of the series. The pieces were arranged in a stack in the sample container with a common cylindrical axis aligned with the applied magnetic field. The measured $\chi_{\text{eff}}$ as a function of $\gamma_p$ for these aligned particles is shown in Fig. 10 for two series, one with $\gamma_g = 20$ and one with $\gamma_g = 11$. $\chi_{\text{eff}}$ initially increases with increasing $\gamma_p$ and levels off for larger $\gamma_p$.

FIG. 8. Effective susceptibility $\chi_{\text{eff}}$ as a function of packing fraction $\phi$ for spherical particles ($\gamma_p = 1$). Squares: dry granular material, $\gamma_g = 11$. Diamonds: suspension, sample aspect ratio $\gamma_g = 11$. Circles: suspension, $\gamma_g = 2.5$. Suspensions exhibit a slightly smaller $\chi_{\text{eff}}$ than dry granular materials. Lines: fit of Eq. 8 for $\gamma_g = 11$ (solid line) and $\gamma_g = 2.5$ (dashed line), where fit parameters are obtained from simultaneously fitting data of Figs. 8, 9, and 10. $\phi_c = 74\%$ (dashed-dotted line) and the measured liquid-solid transition $\phi_c = 40.7\%$ (dotted line). The divergence at the liquid-solid transition predicted by Eq. 4 is not observed in the measurements.

FIG. 9. Effective susceptibility $\chi_{\text{eff}}$ of suspensions as a function of sample sample aspect ratio $\gamma_g$. Solid symbols: spherical particles ($\gamma_p = 1$) at $\phi = 40\%$. Line: model result of Eq. 3 where fit parameters are obtained from simultaneously fitting data of Figs. 8, 9, and 10. Open Symbols: numerical simulation of spherical particles at $\phi = 40\%$ [11].

**B. Variation of $\chi_{\text{eff}}$ with aspect ratios and packing fraction**

Now that we have calibrated the apparatus and established linearity of the response over our measurement range, we now measure the dependence of the effective susceptibility $\chi_{\text{eff}}$ on the sample packing fraction $\phi$, sample aspect ratio $\gamma_g$, and particle aspect ratio $\gamma_p$, to test and fit the model predictions of Eq. 3.

Figure 5 shows how $\chi_{\text{eff}}$ varies with packing fraction $\phi$ for two series of suspensions and one of dry granular materials with spherical particles ($\gamma_p = 1$), and sample aspect ratios $\gamma_g = 2.5$ and 11. Data at $\gamma_g = 2.5$ have been shifted upwards by a factor of 1.4 according to the calibration in Fig. 4. Measurements are made at packing fractions up to the liquid-solid transition $\phi_c = 40.7\pm0.3\%$ for the suspension, defined as the lowest packing fraction where a non-zero yield stress is measured. Measurements of the yield stress for these samples were reported in a previous paper [2]. For each series, $\chi_{\text{eff}}$ increases with increasing $\phi$. On average, $\chi_{\text{eff}}$ of dry granular materials is higher than that of suspensions by 11% at the same $\phi$.

Figure 6 shows the effective susceptibility $\chi_{\text{eff}}$ as a function of sample aspect ratio $\gamma_g$, for spherical particles ($\gamma_p = 1$) and $\phi = 40\%$. To vary $\gamma_g$ and satisfy the condition $L_s < L < L_p$, the sample diameter $d$ had to be varied along with the length. We use $d = 10.2 \pm 0.1$ mm for $\gamma_g < 15$, $d = 7.1 \pm 0.1$ mm for $\gamma_g = 19$, and $d = 3.7 \pm 0.1$ mm for $\gamma_g > 22$. Measurements for $\gamma_g < 5$ have been adjusted upward by 40% according to the calibration in Fig. 4. At small $\gamma_g$, $\chi_{\text{eff}}$ increases with $\gamma_g$, and reaches a plateau for $\gamma_g \gtrsim 10$. For comparison to previous work in Fig. 3, we show numerical simulation results of randomly packed spherical particles at $\phi = 40\%$ [11] (Bjork et al. reported demagnetization factors $D$ [11], which we converted to $\chi_{\text{eff}} = \phi/D$. Their aspect ratio was defined as the inverse of our aspect ratio definition.). The simulation data follow the same trend as ours.
FIG. 10. Effective susceptibility $\chi_{eff}$ of suspensions of cylindrical rods forced to be aligned with the applied magnetic field as a function of particle aspect ratio $\gamma_p$, with $\phi \approx 100\%$. Values of $\gamma_g$ are given in the legend. Lines: model result of Eq. 8 for $\gamma_g = 11$ (dashed line) and $\gamma_g = 20$ (solid line), where fit parameters are obtained from simultaneously fitting data of Figs. 8, 9, and 10. The simultaneous fits here and in Figs. 8 and 9 confirm the validity of Eq. 8 within a root-mean-square difference of 17%.

C. Testing the models for $\chi_{eff}$

The measurements of $\chi_{eff}$ presented in Sec. III over a wide range of packing fraction $\phi$, sample aspect ratio $\gamma_g$, and particle aspect ratio $\gamma_p$ now allow us to test the model of Eq. 8. In Eq. 8 the demagnetization factors $D_g$ and $D_p$ are unspecified functions of sample and particle geometry, respectively. While calculations have been made of $D_g$ for some shapes [11], we are not aware of any model for $D_p$. To fit parameters, we assume that both $D_g$ and $D_p$ follow power laws of the form $D = A\gamma^n$ as shown in Fig. 8 for single-piece solid magnets [15]. Inserting this forms into Eq. 8 with different fit parameters for $D_g$ and $D_p$ yields our fit function

$$\chi_{eff} = \frac{\phi}{A_g\gamma_g^n(1-\phi) + A_p\gamma_p^n\phi}.$$  

We simultaneously least-squares fit all our suspension data in Figs. 8, 9, and 10 to Eq. 8 to obtain the fit parameters $A_g$, $n_g$, $A_p$, and $n_p$. Input error bars were adjusted to 17% to obtain a reduced Chi-squared of 1 from the fit. This indicates the model matches the data within a root-mean-square difference of 17%. Plots of Eq. 8 with these fit parameters are shown in Figs. 8, 9, and 10 where it is seen that the model captures the trends of $\chi_{eff}$ in $\phi$, $\gamma_g$, and $\gamma_p$, respectively. The corresponding best fit parameters are $A_g = 0.4 \pm 0.1$, $n_g = -1.2 \pm 0.1$, $A_p = 0.16 \pm 0.01$, and $n_p = -4.4 \pm 0.3$. The best fit values of $A_g$ and $n_g$ are consistent with the fit values $A = 0.31 \pm 0.01$ and $n = -1.12 \pm 0.02$ from the data of Chen et al. [15] in Fig. 3 confirming that $D_g$ in Eq. 8 is consistent with the demagnetization factor $D$ of individual particles [11].

To compare with the prediction of Martin et al. [12], we plot Eq. 4 as the dashed-dotted line in Fig. 8 with $\phi_c = 74\%$, the value used by Martin et al. [12]. The result using this value of $\phi_c$ happens to match well with our data with sample aspect ratio $\gamma_g = 11$. However, no sample aspect ratio dependence was prescribed in Eq. 4 and the model does not fit well to data at $\gamma_g = 2.5$. Furthermore, the value of $\phi_c$ suggested by Martin et al. [12] is unphysically large for a liquid-solid transition of a random arrangement of particles, where the particles just barely touch each other, which was the physical meaning of $\phi_c$ in Martin et al. [12]. Our suspensions have a liquid solid transition at $\phi_c = 40.7\%$, measured as the lowest packing fraction where the samples exhibit a non-zero yield stress like a solid [7]. To test the physical intent of that model, we plot Eq. 4 with $\phi_c = 40.7\%$ as the dotted line in Fig. 8. This prediction greatly overestimates our measurements, which do not exhibit the divergence at $\phi_c$ of the prediction. The lack of an observed divergence in $\chi_{eff}$ in the approach to $\phi_c$ is similar to simulations of dry granular materials [11].

D. Effect of particle misalignment

In the previous section, we tested the model of Eq. 8 for particles aligned with the applied magnetic field, which was an assumption of the model of Skomski et al. [3]. However, this is not a very practical case, as real suspensions of asperical particles tend to have randomly arranged and oriented particles. To characterize how particle misalignment affects $\chi_{eff}$, we made suspensions of cylindrical particles of various particle aspect ratios. We purchased iron wire (Goodfellow) and cut it to make
cylindrical particles with different particle aspect ratios \( \gamma_p \). To obtain samples with enough particles to avoid significant finite size effects, while minimizing the number of cuts we needed to make, we used different wire diameters of 0.25, 0.5, and 1 mm, for samples with mean particle aspect ratio \( \gamma_p > 10, 5 \leq \gamma_p < 10, \) and \( \gamma_p < 5, \) respectively. For samples of aspect ratio \( \gamma_p = 4.1 \) and length \( L = 42 \text{ mm} \), this results in the ratio of sample diameter to cylinder diameter between 17 and 9, and the ratio of sample diameter to mean particle length between 4.1 and 2.5, which is a range where the value of the packing fraction \( \phi_c \) of the liquid-solid transition is within 4% of the infinite-size system limit [16, 17]. Effects of confinement on alignment are also presumed to be small in this system-size range. For example, in this range the partial particle alignment from this confinement changes the bulk rheology by less than 3% [17], but to our knowledge the effect on \( \chi_{\text{eff}} \) from this confinement has not been characterized.

Before we compare \( \chi_{\text{eff}} \) for particles of different aspect ratio \( \gamma_p \), we first identify a meaningful packing fraction criteria for comparison. Since the packing fraction \( \phi_c \) of the liquid-solid transition varies with \( \gamma_p \) [16, 17], it would not be meaningful to compare at the same absolute packing fraction. Rather, we chose to measure \( \chi_{\text{eff}} \) at a fixed relative packing fraction \( \phi/\phi_c \) near the liquid-solid transition to determine the maximum \( \chi_{\text{eff}} \) we would expect to obtain in the liquid phase for each \( \gamma_p \). Assuming \( \chi_{\text{eff}} \) increases monotonically with \( \phi \) up to \( \phi_c \) for any particle shape as seen for spheres in Fig. 8 this would be the packing fraction where \( \chi_{\text{eff}} \) is maximized for each particle shape, and any lower value of \( \chi_{\text{eff}} \) could be obtained by tuning the packing fraction down to an appropriate value. \( \phi_c \) was measured for each \( \gamma_p \) by observing the change in surface reflectivity as the particles poked through the liquid-air interface of the suspension when \( \phi > \phi_c \) [18]. This transition is sharp and easily observed, allowing us to measure it with an uncertainty on \( \phi_c \) of \( \pm 1\% \). \( \phi_c \) is plotted as a function of particle aspect ratio \( \gamma_p \) in Fig. 11 The horizontal error bars indicate the standard deviation of particle aspect ratios due to the variation in cut particle lengths. The measured \( \phi_c \) decreases with increasing \( \gamma_p \) for cylinders, consistent with previous results [19]. For later input into models, a power law is fit to \( \phi_c \) for cylinders, yielding \( \phi_c = 0.62 \gamma_p^{-0.22} \). For comparison we also plot \( \phi_c \) for the spherical particles used in earlier sections in Fig. 11. The value of \( \phi_c \) for the spheres for not follow the same trend as the cylinders, not only because of the particle shape, but also the different material source may subtly affect interparticle interactions that can have a significant affect on \( \phi_c \) [20].

Since sample preparation procedures can affect the alignment of particles, we also characterize the tendency for the particles to align based on different shaking procedures after the sample was loaded into the cell, but before the magnetic field was applied. We use a suspension with sample aspect ratio \( \gamma_p = 4.1 \), sample length \( L = 42 \text{ mm} \), and particle aspect ratio \( \gamma_p = 5.3 \), at a packing fraction \( \phi/\phi_c = 1.02 \). This packing fraction is just barely resolvable to be above the liquid-solid transition, so we can observe the alignment of the particles as they poke out the liquid-air interface. In one case, samples were shaken along the axis of the cylindrical tube to partly align the particles with the external magnetic field. Open diamonds: samples were shaken using a vortex mixer, resulting in a more random alignment. Pictures of the samples for the 2 shaking procedures are shown in panels (b) and (c), respectively, where the applied magnetic field is aligned in the vertical direction.

![FIG. 12. (a) Probability distribution of particle alignment angles \( \beta \) relative to the applied magnetic field, for samples with length \( L = 42 \text{ mm} \), sample aspect ratio \( \gamma_p = 4.1 \), particle aspect ratio \( \gamma_p = 5.3 \), and packing fraction \( \phi/\phi_c = 1.02 \). Solid squares: samples were shaken along the axis of the cylindrical tube to partly align the particles with the external magnetic field. Open diamonds: samples were shaken using a vortex mixer, resulting in a more random alignment. Pictures of the samples for the 2 shaking procedures are shown in panels (b) and (c), respectively, where the applied magnetic field is aligned in the vertical direction.](image)
quantitative estimate of the average vector component of alignment \( \cos(\beta) \) is also 20\% higher for the particles shaken along the cylinder axis then those shaken by the vortex mixer. It suggests, at least in the ballpark, the decrease of \( \chi_{\text{eff}} \) in Fig. 13 may be associated with the change in particle alignment for these two samples of randomly arranged particles. However, extrapolating this simple estimate does not reach the model of Eq. 8 which suggests that much better alignment would be needed to reach that regime than is likely to be obtained in suspensions with even partially randomly arranged particles, regardless of shaking or other procedures used to get a preferential alignment.

While there is little trend in \( \chi_{\text{eff}} \) over the range of \( \gamma_p \) measured in Fig. 13, it is notable that \( \chi_{\text{eff}} \) exhibits a local maximum in \( \gamma_p \). In contrast, Eq. 8 predicts \( \chi_{\text{eff}} \) to be a monotonically increasing function of \( \gamma_p \) (as seen in Fig. 10). This local decrease in \( \chi_{\text{eff}} \) with \( \gamma_p \) is not due to the different wire diameters used, as in the range 5 \( \leq \gamma_p < 10 \) where \( \chi_{\text{eff}} \) decreased, the same diameter wires were used. Similarly, finite-size effects cannot explain the peak, as the number of particles is decreasing over the same range of \( \gamma_p \), which would only be expected to produce more alignment and a larger \( \chi_{\text{eff}} \), in contradiction to the trend observed in \( \chi_{\text{eff}} \). It could also be proposed that the local peak in \( \chi_{\text{eff}}(\gamma_p) \) could be due to a competition between the increasing \( \chi_{\text{eff}} \) in Eq. 8 and the decreasing \( \phi_c \) with \( \gamma_p \). However, as shown in Fig. 13 the model of Eq. 8 still has no local maximum in this parameter range— even when accounting for this decreasing \( \phi_c \) with \( \gamma_p \). This insensitivity to \( \phi \) in the model is apparent in the limit of large \( \gamma_p \) of Eq. 8 which becomes \( \chi_{\text{eff}} \approx 1/\gamma_p \), independent of \( \phi \). The cause of this local maximum in \( \chi_{\text{eff}}(\gamma_p) \) remains unknown.

The data in Fig. 13 were taken at \( \phi/\phi_c = 1.02 \), corresponding to a jammed state where particles were not free to realign in the applied magnetic field. If instead particles were at a lower packing fraction in a liquid state, they might be expected to be able to more freely and better align with the applied magnetic field to reach the higher \( \chi_{\text{eff}} \) predicted by Skomski et al. 1. To test this hypothesis, we measured \( \chi_{\text{eff}} \) as a function of packing fraction \( \phi \), for 0.5 mm diameter wire cut to length 3.2 mm with a standard deviation of 0.6 mm to obtain a particle aspect ratio \( \gamma_p = 6.3 \pm 1.3 \), near the peak found in Fig. 13. We started with a sample aspect ratio of \( \gamma_p = 4.1 \pm 0.3 \) at \( \phi = 0.42 \) in a 10.2 mm diameter tube, and diluted the sample with more liquid to increase \( \phi \). The sample aspect ratio decreased to 3.8 as the liquid-solid transition was crossed as the suspension packed more efficiently, without trapped air. Upon further dilution, the sample aspect ratio increased in inverse proportion to the packing fraction due to the increase in liquid volume. Because the signal was weaker at these lower frequencies, the calibration of \( \epsilon_{\text{noise}} \) was done with more precision by measuring induced voltage separately before each data point with the current source outputting at the frequency and applied current of the data point but without a sample.
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animals could be approximately applied to other shapes, in particular cylinders. This fit yields Dφ = 0.4γ−1.2, con-
sistent with values obtained for single-piece solids over a smaller parameter range [12], and Dp = 0.16γ−4.4 to character-
ize the particle aspect ratio dependence for the first time, to our knowledge. However, for non-spherical particles randomly oriented in suspensions, the model prediction overestimates the measurements by a factor of 4 for γg = 4.1. As a result of these lower values of χeff for randomly aligned cylinders, we find χeff to be only 20% higher for aspect ratio γ = 6.3 cylinders than spheres. This effect from particle misalignment remains to be included in models. The largest effective susceptibility we found was χeff = 4.8 for cylinders of particle aspect ratio γp = 6.3 and sample aspect ratio γg = 4.1. We also observed that χeff(γp) displays a local maximum at γp = 4 for γg = 4.1. This feature is unexpected, as it was predicted that the maximum χeff would increase monotonically with γp [12]. Another prediction that χeff would diverge to approach the material susceptibility χ at the liquid-solid transition φs [12] fails dramatically, as we observe only χeff ≈ χ in the limit of this transition. This failure to achieve the predicted χeff approaching χ for large packing fraction and/or large aspect ratio may limit applications of magnetic suspensions, as χeff ≈ χ would have allowed for much stronger magnetic responses of suspensions, comparable to ferromagnetic materials. Nonetheless, we do find a significant range of tunable magnetic properties of magnetic suspensions up to χeff ≈ 4 for spherical particles, several orders-of-magnitude stronger than other paramagnetic fluids, which typically have χ in the range of 10−9 to 10−4. The linearity of the magnetic response without hysteresis, like paramagnetic materials, can also be desirable for simple control.

IV. CONCLUSIONS

In this paper, we reported measurements of the effective magnetic susceptibility χeff of suspensions as a function of packing fraction φ, sample aspect ratio γ, and particle aspect ratio γp. When particles are aligned with the applied magnetic field, the model of Skomski et al. [1] can be fit with power laws for the demagnetization factors Dφ and Dp describing the aspect ratio dependence of the sample and particles, respectively, in the form of Eq. 8 with a root-mean-square difference of 17%. This was done by simultaneously varying the three model parameters over the range 0 ≤ φ ≤ 40.7% (up to the liquid-solid transition φs), 2.5 ≤ γ ≤ 32, and 1 ≤ γp ≤ 20. This indicates the model which was originally derived for ellipsoids can be approximately applied to other shapes, in particular cylinders. This fit yields Dφ = 0.4γ−1.2, consistent with values obtained for single-piece solids over a smaller parameter range [12], and Dp = 0.16γ−4.4 to characterize the particle aspect ratio dependence for the first time, to our knowledge. However, for non-spherical particles randomly oriented in suspensions, the model prediction overestimates the measurements by a factor of 4 for γg = 4.1. As a result of these lower values of χeff for randomly aligned cylinders, we find χeff to be only 20% higher for aspect ratio γ = 6.3 cylinders than spheres. This effect from particle misalignment remains to be included in models. The largest effective susceptibility we found was χeff = 4.8 for cylinders of particle aspect ratio γp = 6.3 and sample aspect ratio γg = 4.1. We also observed that χeff(γp) displays a local maximum at γp = 4 for γg = 4.1. This feature is unexpected, as it was predicted that the maximum χeff would increase monotonically with γp [12]. Another prediction that χeff would diverge to approach the material susceptibility χ at the liquid-solid transition φs [12] fails dramatically, as we observe only χeff ≈ χ in the limit of this transition.

This failure to achieve the predicted χeff approaching χ for large packing fraction and/or large aspect ratio may limit applications of magnetic suspensions, as χeff ≈ χ would have allowed for much stronger magnetic responses of suspensions, comparable to ferromagnetic materials. Nonetheless, we do find a significant range of tunable magnetic properties of magnetic suspensions up to χeff ≈ 4 for spherical particles, several orders-of-magnitude stronger than other paramagnetic fluids, which typically have χ in the range of 10−9 to 10−4. The linearity of the magnetic response without hysteresis, like paramagnetic materials, can also be desirable for simple control.

The plateau where χeff was independent of frequency occurred for f < 200 Hz for these cylinders, so the reported χeff was obtained from a weighted average of data in that range. Values of χeff for these particle aspect ratio γp = 6.3 cylinders are shown as a function of packing fraction φ in Fig. 14. We only report χeff for samples shaken in the vortex mixer, as samples shaken vertically to intention-
ally align particles showed an increase in χeff of typically 20%, as found in Fig. 13. For φ ≥ 0.33, the samples had length L < 42 mm, so measured χeff values were scaled up by a factor of 1.4 according to the calibration in Sec. IV. D. χeff increases with φ for cylinders as it does for spheres for φ < φc. We do find a decrease in χeff as φ increases above φc, as expected due to the inability of particles to rearrange for φ > φc. The prediction of Eq. 8 for aspect ratio γp = 6.3 is shown as the solid line in Fig. 14. The prediction is again well above the data, by a factor of 3 or more. A correction for the variation of sample aspect ratio γg from the dilution according to Eq. 8 would not increase χeff by more than 10% for any data point, not nearly enough to match the prediction shown in Fig. 14. For comparison, we also plot the prediction of Eq. 8 for aspect ratio 1 as the dotted line in Fig. 14. The aspect ratio γp = 6.3 particles do have a slightly higher χeff than spheres, and reach up to χeff = 4.8 at the highest packing fraction of the liquid state (φ = 0.39). However, the disagreement with prediction confirms that even in the liquid state, the larger χeff predicted by Skomski et al. [1] is not realized, due to the random arrangement and orientation of particles in suspension which produces a strong demagnetization effect even for large particle aspect ratios.
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