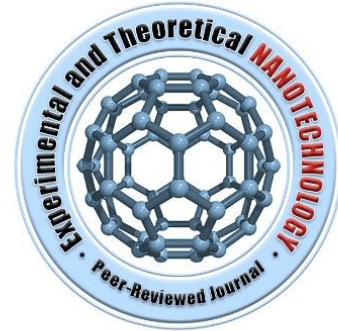# Effect of particle size distribution on the magnetization of ferrofluid

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The effect of particle size distribution on the initial susceptibility of ferrofluids was studied. Magnetization and initial susceptibility were calculated for Uniform, Lognormal and Gaussian distributions. Using statistical mechanics, the magnetization and initial susceptibility of a dimer model were written. Dimer model consists of *N* particles, each particle interacts only with one adjacent particle. The system is exposed to an external magnetic field so the total energy of the system is the sum of the dipole-dipole interaction energy and the particles-field interaction energy. After writing the magnetization and the initial susceptibility it was multiplied by the particle size distribution at two different values of standard deviation. Using Mathematica, the integration over diameter was evaluated. Magnetization curve, Curie-Weiss law and initial susceptibility versus temperature at high fields were investigated for all of the three size distributions. The results were compared with experimental values and we found that Gaussian distribution was the best.

Keywords: Magnetic Susceptibility, Ferromagnetic Materials, Magnetization

1. INTRODUCTION

Magnetic fluids have many applications in medical and industrial areas [1-3], which made the study of these fluids an important and evolving field. Ferrofluid is a liquid that becomes magnetized in the presence of a magnetic field. It consists of magnetic nanoparticles suspended by Brownian motion, which is the random motion of small particles in a fluid; these magnetic particles are surrounded by a nonmagnetic surfactant to prevent agglomeration. Defining these fluids as colloidal suspension comes from the metallic or metallic oxide magnetic particles suspended in a carrier liquid. To form this suspension, three forces must be balanced. These forces are gravitational force, dipole- dipole magnetic force and van der Waals forces. Gravitational force is balanced by Brownian motion. As gravitational force tries to precipitate the magnetic particles in the bottom of the fluid, the Brownian motion of the nanoparticles keeps them suspended.

Magnetic force increases by the increment of the size of the magnetic particles and the concentration of the magnetic fluid. To reduce the magnetic forces, the fluid is diluted and the size of the particle is reduced to nano size. Van der Waals force, which is attractive or repulsive force between molecules, is balanced by an electrostatic force or by steric stabilization. Electrostatic force occurs by inducing positive or negative charge on the magnetic particles, which will cause repulsion. Steric repulsion formed by adding a surfactant to prevent agglomeration [2]. In ferrofluids, each magnetic particle carries a magnetic moment. A proper external magnetic field will affect these moments and enhance many magnetic properties to show up and become easier to be detected and studied.

Magnetic properties of ferrofluids can't be analyzed completely without knowing the size and the size distribution of the magnetic particles in the fluid. Ferromagnetic materials are divided into regions called domains. Each domain contains millions of magnetic moments pointing in the same direction. If the material consists of very small grains, the number of domains in each grain will decrease as its size decreases. In ferrofluids, the grains are in nano size so they contain only one domain, for that they are called single domain magnetic particles. As the grain size continues to decrease, they become superparamagnetic particles with specific magnetic properties [4]. Each particle (grain) contains many magnetic moments arranged in the same direction. The magnetic moment is related to the size of the particle by the relation $\mu = M_s V$, where μ is the magnetic moment, M_s is the saturation magnetization, and V is the volume of the particle.

Studying the effect of particle size in ferromagnetic materials has many great applications. One of the very promising applications is in biomedicine [5-8], since the size ranges from few nanometers to tens of nanometers, i.e., the size of magnetic particle is comparable to the size of biological entities, as an example, a virus (20-450 nm), protein (5-50 nm) and gene (2 nm wide to 10 nm long) [9]. So, one clear application of these magnetic particles is that they can penetrate these biological entities, which can be controlled by external magnetic field which is important in drug delivery and cell separation [10-12].

Many methods are used to measure the size of these nano particles. Actually each method gives a deferent measurement of the size; however, these differences are very small and depend on the aspects that have been measured. For example, electron micrograph is a method to measure the physical size; it determines the solid contents of the particle. The magnetic size, determined from magnetic data, is smaller than the physical size. Hydrodynamic size is the largest of these sizes because it includes the surfactant; it is obtained from viscosity or light scattering data [13].

Using theory and experiment, many researchers tried to construct the relation between magnetization and initial susceptibility with many factors like the magnetic field direction, inter particle interaction, anisotropy, particle size and size distribution [13-23].

Odeh et al [13] used statistical mechanics to calculate magnetization and initial susceptibility of the dimer model in a dilute ferrofluid. They found that Curie temperature and the ordering temperature depend on the direction of the magnetic field with respect to the dimer chain. Obeidat et al [19] used dimer model to study shape and inter-particles interaction on initial susceptibility. They found that anisotropy plays a major role in determining the magnetic state. There is a great theoretical interest in dipoles of interacting particles, because the dipole- dipole interaction is long range and it depends on the orientation of the magnetic particle. Chantrell et al [16] found that initial susceptibility in the limits of small interaction follow Curie-Weiss law. Ayoub et al [14] measured the initial susceptibility of magnetite ferrofluid experimentally.

They found that the initial susceptibility depends on the packing fraction. Their results indicate that, the nature of the inter-particle coupling is antiferromagnetic like.

Szalai et al [24] used Ruelle's algebraic perturbation theory to calculate the free energy expression at which magnetic properties of ferrofluids can be determined, and their results from perturbation theory as well as from Monte Carlo simulations were compared with those of Ivanov and Kuznetsra [25]. The data were in good agreement regarding the initial susceptibility, and their Monte Carlo simulations showed fair agreement regarding the second order nonlinear magnetic susceptibility. Also, Szalai et al [26-28] extended the work of Lebedev [29], who transformed the theory of mean spherical approximation (MSA), which was used originally by Werthem [30] to obtain an analytical expression for the dielectric constant of dipolar fluids, to magnetic fluids. Later on, Kristof et al [31] applied the MSA on two dimensional ferrofluid to obtain an exact expression for the magnetization of dipolar hard disk.

Popplewell et al [21] studied experimentally the effect of particles interactions on Curie-Weiss behavior of ferrofluid. They found that the ferrofluids exhibit Curie-Weiss type behavior with negative Neel temperature. Effects of particle size on the properties of magnetic fluid were studied by Popplewll et al [4]. They found that shape and size are important factors to take into account when interpreting physical and magnetic measurements. Lognormal distribution was found to give the best fit for the experimental results. Ayoub [15] suggested log- normal distribution to calculate susceptibility; he predicted the peak in susceptibility and the effect of concentration on the temperature of that peek.

Particle size distribution of magnetic particles in ferrofluids was determined in many experimental researches. Woodward et al [23] made a comparison between three methods for determining the particle size distribution. The methods were electron microscopy, magnetic measurements and small angle neutron scattering. The distributions determined are Gaussian and Lognormal. Pshenichnikov et al [22] showed that the particle size distribution of magnetite ferro colloids can be approximated with almost the same accuracy either by the Lognormal or by the Gamma distribution by using magneto-granulometric analysis. Biasi et al [17] used ferromagnetic resonance experiments to determine the particle size distribution. They found that the distribution consists of two terms. The first term of the distribution was the lognormal distribution and the second term was a Lifshitz-Slyozov distribution.

Particle size distribution effects on the ferromagnetic resonance were theoretically investigated by Marin [18]. He assumed that the particles diameters obey Gaussian distribution and show that the resonance line, the resonance field and the line width are strongly affected by particle size and temperature. Theoretical calculations were made by Payet et al [20] using the Debye model including the lognormal particles size distribution to show the influence of particle size on the initial susceptibility.

In this work, we will determine the effect of particle size distribution on magnetic properties of a ferrofluid consists of magnetite particles. These particles have different sizes, with diameters ranges between 3 – 11 nm. Our model consists of N particles. Each particle interacts only with one adjacent particle forming N/2 non-interacting systems. The magnetization was calculated when the external field is parallel and perpendicular to the chain of the particles. In our calculation we will consider the particle size distribution. Three different distributions have been considered: Uniform, Lognormal and Gaussian distributions.

The initial susceptibility was calculated depending on the particle size distribution. The calculations curried out for samples assumed to have different concentrations. Then, the ordering temperature was calculated and compared with experimental data obtained from Popplewell et al [21] and Obeidat et al [32].

2. THEORY

Our system of magnetic particles consists of N identical spherical particles, the chain of these particles are constrained to move in one dimension. Each particle interacts with one adjacent particle, so this assembly is divided into $N/2$ non – interacting systems.
The magnetization for this system is calculated from the equation

$$M = k_B T \frac{\partial}{\partial H} \ln Z_T \tag{1}$$

where the total partition function of this system is given by

$$Z_T = \frac{Z^{N/2}}{\left(\frac{N}{2}\right)!} \tag{2}$$

And the partition function Z for two interacting particles is

$$Z = \int \exp\left[-\frac{E_T}{k_B T}\right] d\Gamma \tag{3}$$

The total energy E_T consists of the interaction energy between the magnetic dipole moments (μ) and the external magnetic field (H) and the interaction energy between two dipoles.
Particle size distribution (PSD), in general, is a function that describes the number of particles as a function of diameter, mass or surface area [33]. In our study we define a function $f(y)dy$ as the fraction of the total magnetic volume having reduced diameter between y and $y + dy$, where $f(y) = N(y)V(y)/V_T$, where $N(y)$ is the number of particles having diameter y, $V(y)$ is the particle volume having diameter y, V_T is the total magnetic volume, $y = D/D_v$ is the reduced diameter, D is the particle diameter, and D_v is the median diameter of the distribution. Many functions are suggested to describe the distribution of particles. Here we are interested in studying three different particle size distributions: Uniform, Gaussian, and Lognormal distributions. Common techniques used to determine particle size distribution are transmission electron microscopy (TEM), magnetic measurements and small angle neutron scattering. There is a small deference in the values of standard deviation and average diameter obtained by these techniques [23].
The uniform distribution function is a delta function given as

$$f(D) = \delta(D - D_v) \tag{4}$$

For the Gaussian distribution the particles don't have the same size any more. The Gaussian distribution function is

$$f(D) = \frac{1}{\sqrt{2\pi}\,\zeta} \exp\left[\frac{-(D-D_v)^2}{2\zeta^2}\right] \tag{5}$$

Here ζ is the standard deviation of D. For log normal particle size distribution we use

$$f(D) = \frac{1}{\sqrt{2\pi}\,\sigma D} \exp\left[\frac{-(\ln D/D_v)^2}{2\sigma^2}\right] \tag{6}$$

where σ is the standard deviation of $\ln D$. By considering particle size distribution function the overall magnetization can be written as a function of the magnetic field

$$M' = \int M(y, H) f(y) dy \tag{7}$$

The initial susceptibility of the system then can be calculated as:

$$\chi = \lim_{H \to 0} \frac{\partial M'}{\partial H} \tag{8}$$

3. RESULTS AND DISCUSSION

Cas1: Parallel field
Consider the external applied field to be parallel to the chain of particles as shown in Fig. 1.
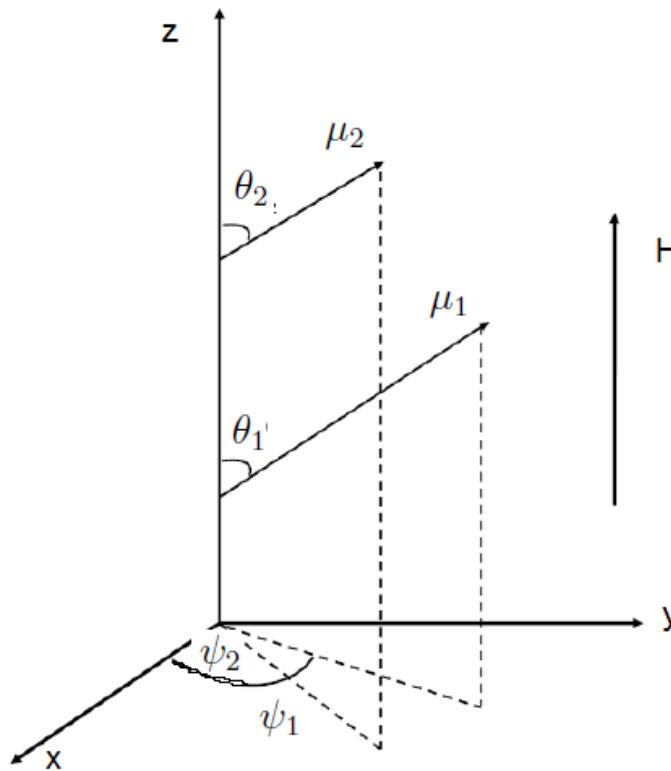

Figure 1 Applied field parallel to the chain of particles.

The field particle energy is given by:

$$E_0 = -\mu_1 H \cos\theta_1 - \mu_2 H \cos\theta_2 \tag{9}$$

And the interaction energy between the two dipoles is

$$E_{int} = \frac{\mu_1 \mu_2}{z^3} g(\theta_1, \theta_2, \psi_1, \psi_2) \tag{10}$$

where $g(\theta_1, \theta_2, \psi_1, \psi_2) = 2\,cos\theta_1\,cos\theta_2 - sin\theta_1\,sin\theta_2\,cos(\psi_1 - \psi_2)$

Assuming that the interaction energy between the dipoles is very small compared to the thermal energy, and then the partition function can be written as:

$$Z = \int_{z_n}^{z_m} \int_0^{2\pi} \int_0^{2\pi} \int_0^{\pi} \int_0^{\pi} \frac{\exp\left[\frac{\mu_1 H}{k_B T}cos\theta_1 + \frac{\mu_2 H}{k_B T}cos\theta_2\right]}{\left\{1 + \frac{\mu_1 \mu_2}{z^3 k_B T} g(\theta_1, \theta_2, \psi_1, \psi_2)\right\}} d\Gamma \tag{11}$$

where $z_n = \frac{D_1+D_2}{2} + 2\delta$, and $z_m = \frac{D_1+D_2}{2\sqrt[3]{4\varepsilon}}$ are the minimum and maximum separation respectively. δ is the thickness of the surfactant and ε is the volumetric packing fraction. Using the modified spherical Bessel functions, the magnetization for this system can be written as:

$$M = \frac{N}{2V_s}\left(\mu_1 \frac{i_1(x_1)i_0(x_2) + \frac{\mu_1\mu_2}{k_B T}\left(\frac{z_m + z_n}{z_m^2 z_n^2}\right)\left[i_0(x_1) - \frac{2}{x_1}i_1(x_1)\right]i_1(x_2)}{i_0(x_1)i_0(x_2) + \frac{\mu_1\mu_2}{k_B T}\left(\frac{z_m + z_n}{z_m^2 z_n^2}\right)i_1(x_1)i_1(x_2)}\right.$$
$$\left. + \mu_2 \frac{i_1(x_2)i_0(x_1) + \frac{\mu_1\mu_2}{k_B T}\left(\frac{z_m + z_n}{z_m^2 z_n^2}\right)\left[i_0(x_2) - \frac{2}{x_1}i_1(x_2)\right]i_1(x_1)}{i_0(x_1)i_0(x_2) + \frac{\mu_1\mu_2}{k_B T}\left(\frac{z_m + z_n}{z_m^2 z_n^2}\right)i_1(x_1)i_1(x_2)}\right) \tag{12}$$

where V_s is the volume of the sample and $x_j = \frac{\mu_j H}{k_B T}$.

The effect of the particle size distribution has been determined by solving equation (4) numerically using the three different types of distributions. In our model we have chosen the minimum diameter of the particles to be 3 nm and the maximum diameter to be 11 nm to maintain the condition $\mu_1 \mu_2 < k_B T\, z^3$. Fig. 2 shows the magnetization curves of the three different distributions for a packing fraction $\varepsilon = 0.05$ and a temperature $T = 300\,K$. The three curves show a similar behavior for high magnetic field and they reach the saturation around 2500 Oersted. However they have different behaviour for low field which indicate different values of the initial susceptibility.
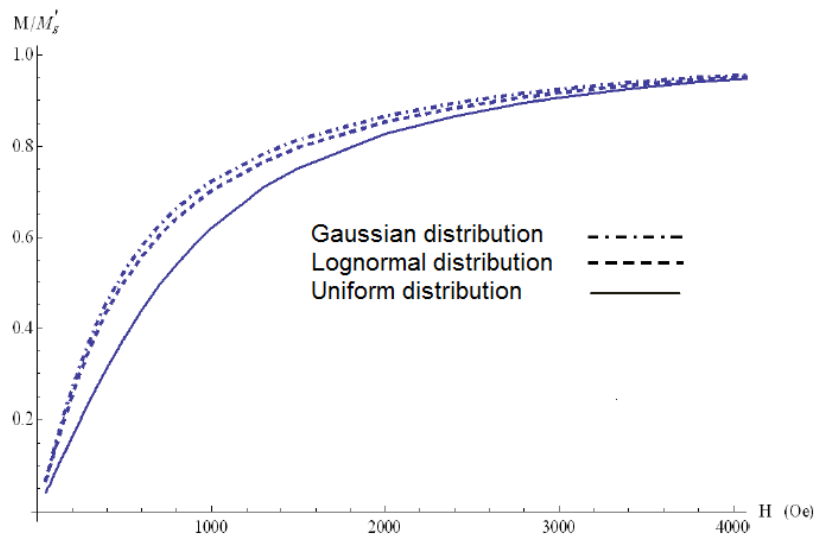
Figure 2 Magnetization curve of uniform, Gaussian and log normal distributions.

Case 2: Perpendicular field
For this case we will consider the external applied field to be perpendicular to the chain of particles as shown in Fig. 3.
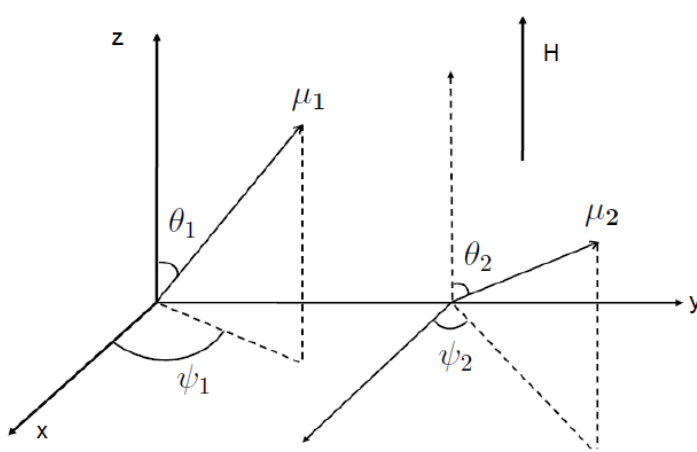

Figure 3 Applied field is perpendicular to the chain of particles.

The total energy of the system is given by

$$E = -\mu_1 H \cos\theta_1 - \mu_2 H \cos\theta_2 - \frac{\mu_1\mu_2}{y^3} f(\theta_1, \theta_2, \psi_1, \psi_2) \tag{13}$$

where $f(\theta_1, \theta_2, \psi_1, \psi_2) = 3 \sin\theta_1 \sin\theta_2 \cos\psi_1 \cos\psi_2 - \cos\theta_1 \cos\theta_2 - \sin\theta_1 \sin\theta_2 \cos(\psi_1 - \psi_2)$

Following the same procedure as for the parallel field the magnetization can be written as

$$M = \frac{N}{2V_s}\left(\mu_1 \frac{2i_1(x_1)i_0(x_2) - \frac{\mu_1\mu_2}{k_BT}\left(\frac{y_m+y_n}{y_m^2 y_n^2}\right)\left[i_0(x_1) - \frac{2}{x_1}i_1(x_1)\right]i_1(x_2)}{2i_0(x_1)i_0(x_2) - \frac{\mu_1\mu_2}{k_BT}\left(\frac{y_m+y_n}{y_m^2 y_n^2}\right)i_1(x_1)i_1(x_2)} \right.$$
$$\left. + \mu_2 \frac{2i_1(x_2)i_0(x_1) - \frac{\mu_1\mu_2}{k_BT}\left(\frac{y_m+y_n}{y_m^2 y_n^2}\right)\left[i_0(x_2) - \frac{2}{x_1}i_1(x_2)\right]i_1(x_1)}{2i_0(x_1)i_0(x_2) - \frac{\mu_1\mu_2}{k_BT}\left(\frac{y_m+y_n}{y_m^2 y_n^2}\right)i_1(x_1)i_1(x_2)} \right) \tag{14}$$

In this equation we have used y to represent the distance between the two particles instead of z since they are laying on the y axis (see Fig. 3).

Fig. 4 shows the magnetization curves for the lognormal distribution at $T = 300\ K$ and a packing fraction of 0.05 for both parallel and perpendicular fields. Both curves show a similar behaviour and a saturation magnetization above 2500 oersted. The initial susceptibility depends on the direction of the magnetic field and that's shown from the behaviour of the magnetization at low fields where each direction has a different path at low fields.
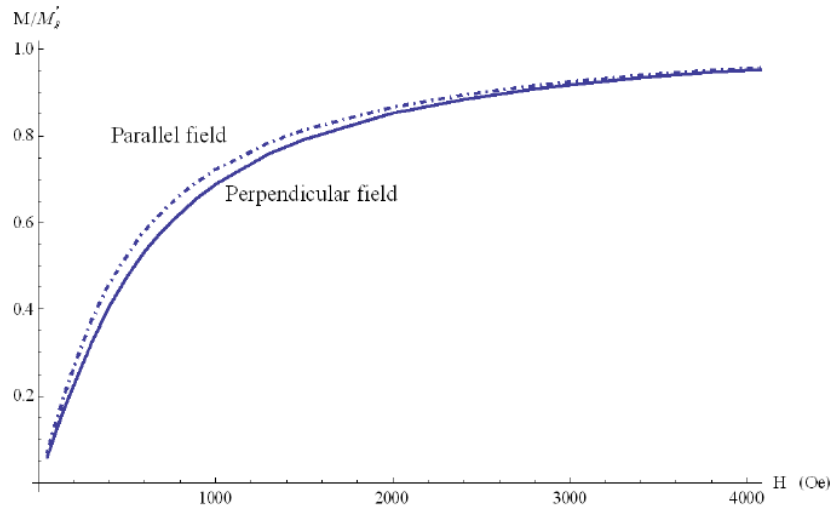
Figure 4 Magnetization curve of the lognormal PSD at $\eta = 0.05$.

The initial susceptibility for this model follows Curie – Weiss law $\chi = \frac{C}{T - T_0}$, where C is the Curie constant and T_0 is the ordering temperature. We found that the material has ferromagnetic behaviour for parallel field while it has an anti-ferromagnetic behaviour for perpendicular field. The ordering temperature has twice the value for parallel field.

Table 1 shows the calculated ordering temperature for Fe_3O_4 when the field is parallel to the chain for different values of the packing fraction ε. In this calculation we have assumed that $D_v = 7.4\ nm$ and $\sigma = 0.4$. The results are compared with experimental data obtained by Popplewell et al [21]. The measurement carried out at a temperature range $220 \sim 450\ K$. The uniform distribution gives the largest error while using lognormal distribution decreases the error. However the Gaussian distribution gives the best results. Moreover, most theoreticians prefer lognormal distribution [4] and suggest that it is more physically acceptable since no values of the diameter are below zero.

Table 1 The ordering temperature for Fe_3O_4 parallel magnetic field.

Packing fraction	Experimental [21]	dimer [32]	trimer [34]	dimer + uniform	dimer + Lognormal	dimer + Gaussian
0.02	10	8.62	12	12.4	12.7	14.2
0.03	19	11.37	19	14.8	15.2	17
0.05	38	13.26	32	18.7	19.2	21.3
0.07	48	15.5	45.4	21.9	22.4	24.4

4. CONCLUSIONS

In this work the effect of particle size distribution on magnetization and initial susceptibility is studied. For dimer model, in solid and in liquid state, the magnetization calculated considering once the Uniform distribution then the Gaussian distribution finally the Lognormal distribution. The magnetization was calculated in two cases, when the applied field is parallel to the chain of assembly and when the field is perpendicular to that chain. For each of the three distributions, the magnetization was affected by the direction of the applied field, at low field there is no

coincidence between the curves drawn for parallel field with that drawn for perpendicular field. And at high field both parallel and perpendicular curves reach the same saturation value. For different particle size distribution, the magnetization curve reaches saturation at the same values. But, before saturation each distribution has a different path. This different path before saturation, indicate that each distribution gives different initial susceptibility. Moreover, initial magnetic susceptibility was calculated depending on these distributions, and then compared with experimental results given in [21]. We found that the calculated results follow Curie-Weiss law. For all distributions, when susceptibility calculated by assuming very small perpendicular field, the ordering temperature was negative and showed anti-ferromagnetic material. By assuming very small parallel field for the same distributions, the ordering temperature obtained was positive indicating that the material has a ferromagnetic behaviour. In addition, we found that T_0 for the parallel is twice that for the perpendicular field. The T_0 obtained from Gaussian distribution is the largest. The Gaussian distribution gives the closest values to the experiment. The Gaussian distribution was better in theoretical calculation because, the amount of large particles involved in calculations using Gaussian distribution are much more than those involved using lognormal distribution, where, the Lognormal distribution has a slow decay in the domain of the large particles. For solid state, the Lognormal and the Gaussian distributions give larger values for T_0 compared with values of liquid state.

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