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# Effect of magnetic anisotropy on the two dimensional dimer model in ferrofluids

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## Abstract

The initial magnetic susceptibility of a two dimensional ferrofluid system has been calculated. Assuming a two body interaction approach, we find that the ordering temperature $T_0$ depends on both of the anisotropy energy of the particles and the direction of the magnetic field. Our calculations illustrate the effect of the particle's size as a sensitive parameter of determining the state of the assembly magnetization.

**Keywords:** Magnetic anisotropy; Ferromagnetism; Antiferromagnetism.
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## 1. Introduction

The magnetic anisotropy of a ferromagnetic fine particle is characterized by its magnetic easy axis. The deviation of the particle magnetization *M* from its easy axis direction (c-axis) measures the magnetic anisotropy of the particle. When a fluid of such particles is subject to an external magnetic field *H*, its magnetic state strongly depends on the nature of the magnetic anisotropy of the particles and on the magnitude and direction of the applied magnetic field. Therefore, many assumptions have been made to explore the relation between *M* and *H* for anisotropic magnetic fluids [1-2]. Stoner *et al* assumed that *M* is parallel to the c-axis if the applied magnetic field is zero [3]. When *H* is applied to such a system of ferrofluid all particles will orient themselves in the direction of *H*. Gruyters [4] studied the two dimensional layers of interacting nanoparticles with random magnetic anisotropy using Monte Carlo technique and found that the random magnetic anisotropy emerges as a new approach to the problem of exchange bias in nanoparticles systems. Franco and Conde [5] studied the effect of the magnetic anisotropy at temperatures above the blocking temperature of uniaxial nanoparticle systems, and they found an influence of anisotropy on the grain size distribution even for monodisperse systems.

Studying the temperature dependence of the initial magnetic susceptibility $\chi$ for a magnetic fluid is a key to classify the magnetic state of the fluid. In the absence of magnetic anisotropy it was found that the Neel temperature increases linearly with increasing magnetic particle concentration in the dilute ferrofluid [6]. For a linear chain of magnetic particles a

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ferromagnetic like state is established when $H$ is parallel to the chain and an anti ferromagnetic like state exists in the perpendicular case [7-8]. Introducing the magnetic anisotropy of the particles showed a significant dependence of the ordering temperature on the anisotropy constant [4, 6-12]. In a two dimensional case, and ignoring the magnetic anisotropy of the particles, it was found that the ordering temperature $T_0$ is always negative, regardless of the field direction [13].

The purpose of the present work is to investigate the effect of particle anisotropy on a dimers magnetic fluid, in the context of the following assumptions:

1. particle-particle interaction (dimer model)
2. the field direction is taken relative to the c-axis (the easy direction) of the magnetic particles
3. Maxwell-Boltzmann statistics is considered in calculating the magnetization, M, of the assembly from which we deduce the initial susceptibility.

There are many types of magnetic anisotropy. We are concerned with two types, namely, the crystal anisotropy or magneto crystalline anisotropy and shape anisotropy. The main source to the crystal anisotropy is spin-orbit coupling. The anisotropic energy of this type can be expressed as, $H_a = KV\sin^2\beta$, where $K$ is the uniaxial anisotropy constant, $V$ is the particle volume, and $\beta$ is the angle between the direction of the magnetic moment of the particle and its easy axis.

In a ferrofluid, shape anisotropy is more dominant than crystalline anisotropy, and shape anisotropy arises because of the shape elongation of the fine nanoparticles. In elongated particles the easy axes are directed along the long axes of the particles [14]. The shape anisotropy energy is given by

$$H_a = K_1\sin^2\beta + K_2\sin^4\beta + .... \tag{1}$$

Where $K_1$ and $K_2$ are the first and the second order anisotropy constants, respectively. Since it is mathematically easier to deal with spherical particles than elongated particles, so in our model we will consider particle-particle interaction in two dimensions, and assume all the particles are spheres with crystal anisotropy that has the same value as the shape anisotropy. Therefore, only $K_1$ exists (we will refer to it as $K$).

## 2. Theoretical model

Let us assume that our assembly consists of N particles, and each system consists of two particles. To calculate the total magnetization $M$ of the assembly we have to calculate the total partition function $Z_T$ which is equal to $Z_T = \frac{(Z)^{N/2}}{(N/2)!}$, where $Z$ is the two particle partition function and is equal to $Z = \int e^{\frac{-H_T}{kT}} d\Gamma$. Here $H_T$ is the total Hamiltonian energy of the assembly and $\Gamma$ is the volume phase space. In this assembly there are $N/2$ pairs of particles. The total Hamiltonian of the system is given by

$$H_T = H_{\text{int}} + H_0 + H_a \text{ ,} \tag{2}$$

where $H_{\text{int}}$ is the dipole-dipole magnetic interactions, $H_0$ is the magnetic interaction with the external magnetic field $\vec{H}$ and $H_a$ is the anisotropy energy of the particle. These field interactions are, respectively, given by:

$$H_{\text{int.}} = \frac{\vec{\mu}.\vec{\mu}'}{r^3} - \frac{3(\vec{\mu}.\vec{r})(\vec{\mu}'.\vec{r})}{r^5}$$
$$H_0 = -\vec{\mu}.\vec{H} - \vec{\mu}'.\vec{H}$$
$$H_a = KV\sin^2\beta$$

Where $r$ is the separation between the particles, $\vec{\mu}$ and $\vec{\mu}'$ are the magnetic moments of the two particles with same magnitude and different directions, $K$ is the uniaxial anisotropy constant, $V$ is the volume of the particle, and $\beta$ is the angle between the direction of the magnetic moment and the easy axis.

Before we calculate *Z* let us address the assumptions we will consider to simplify the calculation and also give their validity. In the first assumption, we will consider the interaction energy between the dipoles to be very small compared with the thermal agitation energy ($\frac{\mu^2}{kTR_i^3} << 1$), where $R_i$ is the minimum distance between the particles in the system. Secondly, we will assume that the anisotropic energy is small compared with the thermal agitation energy$\left(\frac{KV}{kT} << 1\right)$, this is the case for a dilute system, i.e., the packing fraction, $\varepsilon \le 0.07$, above this critical value, one cannot ignore the effect of viscosity. Finally, the applied magnetic field is small. All these assumptions are valid since we are working at room temperature.

Having addressed the essential parts of our theoretical approach, we have calculated the total magnetization and the initial susceptibility.

### 2.1 Model Calculation

As for the external magnetic field, two configurations are considered. In the first case we consider the field *H* to be perpendicular to the plane and in the second case we consider H to be in-plane.

CASE I:

Figure (1) shows this configuration. The particles lay in the xy-plane and H is in the z-direction. The partition function for this case is given by $Z = \int e^{\frac{-(H_{\text{int}}+H_0+H_a)}{kT}} d\Gamma$. Referring to figure (1) the following expressions for the energy terms of the Hamiltonian, are given by:

The interaction energy $H_{int}$ is:

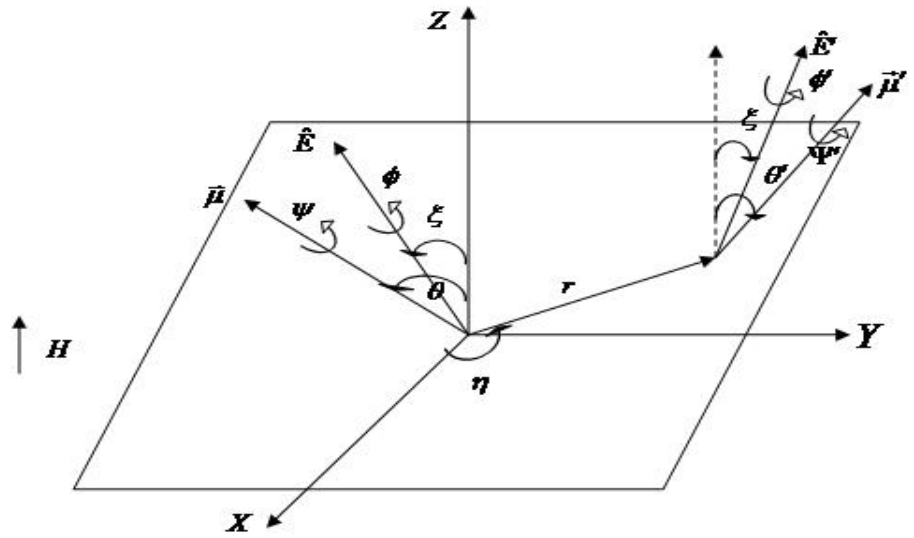


Fig. 1: System of two particles in a plane, the first particle is at the origin, and the second particle at ($r$,η).

Each particle has magnetic dipole moment μ(μ') oriented at angle θ(θ') relative to the z-axis and azimuth al angle ψ(ψ'). $\hat{E}$ is the easy axis oriented at angle ξ relative to the z-axis and azimuth al angle ϕ(ϕ'). The applied field H is parallel to the z-axis

$$H_{\text{int}} = \frac{-\mu^2}{r^3} g(\theta,\theta',\psi,\psi',\eta), \tag{3}$$

Where

$$g(\theta,\theta',\psi,\psi',\eta) = 3\sin\theta\sin\theta'\cos(\eta-\psi)\cos(\eta-\psi') - \sin\theta\sin\theta'\cos(\psi-\psi') - \cos\theta\cos\theta'$$

The anisotropic energy term $H_a$ is:

$$H_a = 2KV - KVJ(\theta,\theta',\xi,\psi,\psi',\phi,\phi'), \tag{4}$$

Where $J(\theta,\theta',\xi,\psi,\psi',\phi,\phi') = \cos^2\beta + \cos^2\beta'$

Finally, the magnetic energy due to the interaction with the magnetic field is equal to

$$H_0 = -\mu H(\cos\theta + \cos\theta') \tag{5}$$

Therefore, the total Hamiltonian can be written as

$$H_T = \frac{-\mu^2}{r^3} g(\theta,\theta',\psi,\psi',\eta) + 2KV - KVJ(\theta,\theta',\xi,\psi,\psi',\phi,\phi') - \mu H(\cos\theta + \cos\theta') \tag{6}$$

All symbols are defined in the caption of figure (1).

Inserting $H_T$ in *Z*, we obtain,

$$Z = e^{\frac{-2KV}{kT}} \int_{r_i}^{r_o} \int_0^{\pi} \int_0^{\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \exp\left(\frac{\mu H}{kT}(\cos\theta + \cos\theta') + \frac{\mu^2}{kTr^3} g + \frac{KV}{kT} J\right) d\Gamma , \tag{7}$$

Where $d\Gamma = d\eta d\psi d\psi' d\phi d\phi' \sin\theta d\theta \sin\theta' d\theta' r dr$ .

where $r_i$ as the minimum separation that depends on the mean diameter of the particle, *D*, as well as the surfactant layer, $\delta$, through the relation $r_i = D + 2\delta$, and $r_0$ as the maximum separation $r_0 = D/(4\varepsilon)^{1/3}$, where $\varepsilon$ is the volumetric packing fraction.

Within the frame of assumptions mentioned above the partition function reduces to

$$Z = e^{\frac{-2KV}{kT}} \int_{r_i}^{r_o} \int_0^{\pi} \int_0^{\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \exp\left(\frac{\mu H}{kT}(\cos\theta + \cos\theta')\right) \left[1 + \left(\frac{\mu^2}{kTr^3} g + \frac{KV}{kT} J\right) + \frac{1}{2}\left(\frac{\mu^2}{kTr^3} g + \frac{KV}{kT} J\right)^2\right] d\Gamma \tag{8}$$

The total magnetization of the system and the initial susceptibility are given by:

$$\mathrm{M} = kT \frac{\partial \ln Z_T}{\partial \mathrm{H}} = \frac{NkT}{2Z} \frac{\partial x}{\partial H} \frac{\partial Z}{\partial x}, \tag{9}$$

where $x = \frac{\mu \mathrm{H}}{kT}$. Therefore, $M = \frac{N\mu}{2Z} \frac{\partial Z}{\partial x}$

$$\chi = \lim_{H \to 0} \left(\frac{\partial M}{\partial H}\right) \tag{10}$$

The last equation, after simplifying it using Taylor expansion based on the small values in the denominator, reduces to

$$\chi_{\perp} = \frac{N\mu^2}{3k} \left[ \frac{1}{T + \frac{2C_1}{3C_0} + \frac{C_2}{3C_0} + \frac{C_2}{C_0} \cos^2 \xi} \right] \tag{11}$$

Where

$$C_0 = 64\pi^5 \frac{\mu^2}{k} e^{-2KV/kT} \left(\frac{1}{r_i} - \frac{1}{r_0}\right) \tag{12}$$

$$C_1 = 128\pi^5 \frac{KV}{k} e^{-2KV/kT} (r_0^2 - r_i^2) \tag{13}$$

$$C_2 = \pi^5 \frac{\mu^4}{k^2} e^{-2KV/kT} \left( \frac{1}{r_0^4} - \frac{1}{r_i^4} \right) \tag{14}$$

CASE II:

Figure (2) shows the configuration of this case, for which the external magnetic field is in the z-direction while the assembly is in the *x*-*z* plane.
The terms in the total energy of the system can be written as

$$H_0 = -\mu H(\cos\theta + \cos\theta')$$
$$H_{\text{int}} = \frac{-\mu^2}{r^3} g(\theta, \theta', \psi, \psi', \eta)$$
$$H_a = 2KV - KVJ(\theta, \theta', \zeta, \psi, \psi', \phi, \phi'),$$

where all symbols are defined in the caption of figure (2).

The initial magnetic susceptibility can be calculated to be:

$$\chi_{\parallel} = \frac{N\mu^2}{3k} \left[ \frac{1}{T - \frac{C_1}{3C_0} + \frac{C_2}{3C_0} + \frac{C_2}{C_0}\cos^2\xi} \right] \tag{15}$$

where $C_0$, $C_1$, $C_2$ are given in equations 12-14.

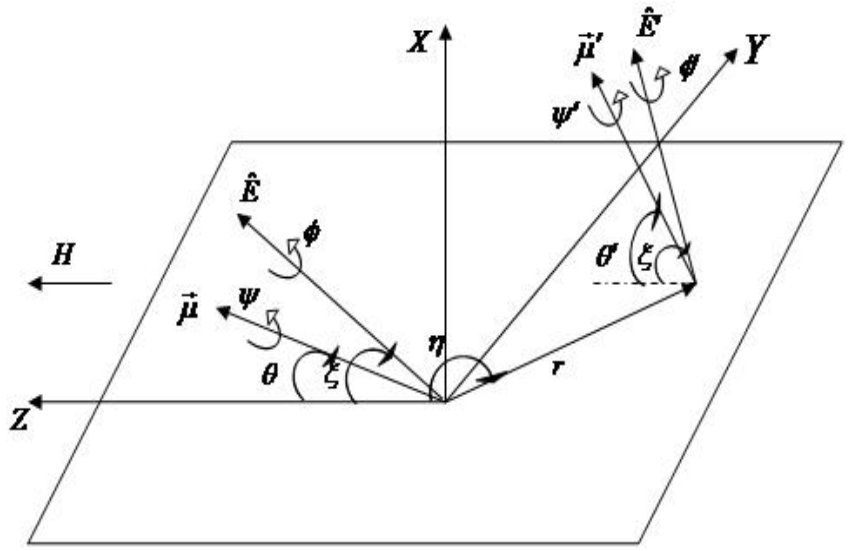


Fig. 2: The applied field H is parallel to the plane. All other symbols are defined in Figure 1.

### 3. Discussion

Equations 11 and 15 give the magnetic susceptibility for the two distinct configurations considered above. For the perpendicular case, substituting the values of $C_0$, $C_1$ and $C_2$ in equation 11 we get the ordering temperature $T_0^{\perp}$ for the perpendicular case as:

$$T_0^{\perp} = -\frac{2\mu^2}{3k}\frac{1}{r_0 r_i (r_0 + r_i)} - \frac{2KV}{3k}(1 - 3\cos^2\xi) \tag{16}$$

The first term of the ordering temperature depends on the interaction between the two dipoles and the separation between particles while the second term depends on the uniaxial anisotropy constant and the volume of the particle. In fact the second term was completely absent when the magnetic anisotropy was ignored in many theoretical works [13, 15-20]. Moreover, the particles separation appears as a new parameter in the ordering temperature. It is worth mentioning that for a random distribution of easy axes, the anisotropy has no effect [7,12].

Now, for the parallel case and after substituting the constants $C_0$, $C_1$ and $C_2$ in equation 15, the ordering temperature $T_0^{\parallel}$ can be written as:

$$T_0^{\parallel} = \frac{\mu^2}{3k}\frac{1}{r_0 r_i (r_0 + r_i)} - \frac{2KV}{3k}(1 - 3\cos^2\xi) \tag{17}$$

This has the same form as the perpendicular case except with the sign of first term is negative and doubled. This means that the dominant term will determine the state of magnetization. In the case of the random distribution of the easy axes our result shows that the anisotropy has no effect as was found in the work obeidat et al[7].

For random distribution, our results show that the ordering temperature is K independent, since for random distribution of the easy axis, the average value of $\cos^2\xi$ is 1/3. Therefore, the ordering temperature is negative (antiferromagnetic like behavior) when the applied magnetic field is perpendicular to the sample, and is positive (ferromagnetic like behavior) when the applied magnetic field is parallel to the sample.

The above results have been compared with the work of Popplewell et al [6] and with the theoretical work of [7-8, 21], their results show a negative ordering temperature of magnetite, so we used the $T_0^{\perp}$ expression for comparison to be consistent with the sign. Table (I) shows the experimental results of the ordering temperature for different values of packing fraction. The mean diameter of the particle, $D_v$, is taken to be 7.4 nm and the surfactant layer, $\delta$, is 2 nm.

Table 1: Comparison between experimental results of the ordering temperature versus the packing fraction of $Fe_3O_4$ with our model and other previous models in literature.

| $\varepsilon$ | [i]$T_0^{exp}$ (K) | $T_0$ (K) this work | [ii]$T_0$ (K) | [iii]$T_0$ (K) | [iv]$T_0$ (K) |
|---|---|---|---|---|---|
| 0.01 | 0 | 6 | 6 | 6.3 | 6.25 |
| 0.02 | 10 | 13.7 | 12 | 10.07 | 8.62 |
| 0.03 | 19 | 20.6 | 19 | 13.2 | 11.37 |
| 0.05 | 38 | 34.5 | 32 | 18.55 | 13.26 |
| 0.07 | 48 | 48 | 45.4 | 23.22 | 15.5 |
| 0.08 | 75 | 54.9 | 52 | 25.38 | |

i Popplewell et al, J. Appl. Phys. **64**, 10 (1988)
ii Gharaibeh et al, accepted to Jordan Journal of Physics
iii Chantrell and Wohlfarth, J. Magn, Magn, Mater. **40**, 1 (1983)
iv Obeidat et al, J Supercond Nov Magn, **22**, 805 (2009)

If we ignore the packing fraction of 0.01, we see that the trimer model is a good model for small values, and the current model is in good agreement for dilute ferrofluid $\varepsilon \leq 0.07$ [14]. Also we see that the model of Chantrell et al [21] is very good also for very dilute ferrofluid, since they consider the rotation of the magnetic moments to be in two dimensions as well as the dimer is constrained to move in a plane.

## 4. Conclusion

We have studied the effect of crystal anisotropy in two dimensional ferrofluid assembly consisting of N particles. The ordering temperature $T_0$ was found to depend on the anisotropy energy of the particles for perpendicular and parallel applied field. The particles separation appears as a new parameter in $T_0$. The dipole-dipole term in $T_0$ is negative for the perpendicular field and is positive for the parallel field. Therefore, the magnetic state is very crucial to this parameter and to the anisotropy. Moreover, our model is in good agreement with the experimental results for dilute systems.

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