THE EFFECT OF THE DIRECTION OF THE MAGNETIC FIELD ON THE CURIE TEMPERATURE FOR A TEXTURED DILUTE FERROFLUID

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الخلاصة :

يُعنى هذا البحث بحساب درجة التمغنط وقابلية التمغنط الابتدائية لسلسلة خطية من الجسيمات المغناطيسية في سائل مغناطيسي مخفف بوجود مجال مغناطيسي مواز أو متعامد مع السلسلة . لقد وجدنا أن درجة الحرارة المميزة تكون موجبة في حالة المجال المغناطيسي الموازي للسلسلة وتكون سالبة في حالة المجال المغناطيسي المتعامد مع السلسلة . كما وجدنا أن مقدار درجة الحرارة المميزة في حالة المجال الموازي تكون ضعف تلك في حالة المجال المتعامد .

ABSTRACT

We have calculated the magnetization and the initial susceptibility for a chain structured dilute ferrofluid, when the applied magnetic field is parallel or perpendicular to the chain. We have found that the Curie temperature in the parallel field case is positive while that in the perpendicular field case is negative, and that the magnitude of the Curie temperature in the first case is twice that in the second case.

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1. INTRODUCTION

A ferrofluid is an ultra-stable suspension of fine ferromagnetic or ferrimagnetic particles in a carrier liquid [1]. The particles are coated with a surfactant layer to minimize agglomeration [2]. To study and characterize the magnetic properties of such fine particle systems, two quantitities are of paramount importance, the magnetization, M, and the initial susceptibility, χ_i.

Several experimental studies have been carried out on magnetic fine particle systems, both in the liquid and the solid state [3–6]. Rosman *et al.* [7] have found that, at low concentration, the ferrofluid behaves like small chain aggregates in the presence of an applied magnetic field of 0.1 tesla, where the ferrofluid corresponds to an assembly of independent pairs of particles (dimers) oriented along the field direction. In general the interparticle interactions in a ferrofluid are reflected in the existence and the sign [4] of the Curie temperature T_0 obtained from the Curie–Weiss $(C - W)$ law, $\chi_i = C/(T - T_0)$, where C is Curie constant and T is the absolute temperature. The Curie temperature T_0 could be either negative [4] indicating an anti-ferromagnetic behavior or positive [8] indicating ferromagnetic behavior.

Theoretically the simplest model is that in which the interparticle interactions are negligible and the Langevin theory for uniform size particles is applicable [9]. Introducing the particle size distribution into Langevin theory, we obtain the theory proposed by Chantrell *et al.* [10]. This theory predicts a Curie's law with T_0=0. However it is a good theory for very dilute ferrofluids. The mean field approximation [9] is an improvement on this theory where the interparticle interactions are taken into account in an average manner.

In the present work we carry out calculations for the magnetization and the initial susceptibility of a system of N particles constrained to move on a straight line in a dilute ferrofluid. We will be using the dimer model (DM) for taking into consideration the interparticle interactions where each particle interacts with only one other adjacent particle of the linear assembly. We consider two cases: in the first case the applied magnetic field is parallel to the assembly and in the second case the applied magnetic field is perpendicular to the assembly. These two cases are analyzed in Sections 2.1, 2.2 respectively. In Section 3 we discuss the results and state the conclusions.

2. THEORY

Consider a system of N identical fine magnetic particles each of diameter D that are constrained to move along a straight line of length L along the x-axis. The magnetic moment $\vec{\mu}$ of any particle of the system is free to rotate in three dimensions. In the dimer model the interparticle interaction of the pair is that due to magnetostatic interaction. The interaction energy between two magnetic dipoles is given by

$$E_0 = \frac{\vec{\mu} \cdot \vec{\mu}'}{x^3} - \frac{3(\vec{\mu} \cdot \vec{x})(\vec{\mu}' \cdot \vec{x})}{x^5}$$

where x is the particle separation. In the presence of an applied magnetic field $\vec{H}$, the total magnetic energy, E, of a pair is given by

$$E = E_0 - \vec{\mu} \cdot \vec{H} - \vec{\mu}' \cdot \vec{H}$$

To calculate the magnetization M of the system we use statistical mechanics and start by calculating the partition function Z for a pair of particles:

$$Z = \int_{x_i}^{x_o} dx \int_0^{2\pi} \int_0^{\pi} \sin\theta d\theta d\psi \int_0^{2\pi} \int_0^{\pi} \sin\theta' d\theta' d\psi' e^{-E/kT} \tag{1}$$

where k is the Boltzmann constant, T is the temperature of the system, θ and θ' are the respective angles which $\vec{\mu}$ and $\vec{\mu}'$ make with the direction of $\vec{H}$ and ψ, ψ' are their corresponding azimuthal angles (Figures 1 and 2). $x_i = D + 2\delta$ is the minimum separation between the two particles, δ being the thickness of the surfactant layer; $x_0 = 2D/\eta$ is the maximum separation of the dimers, where $\eta = ND/L$ is the packing fraction in one dimension.

Now in order to calculate the partition function Z, we shall expand the exponential factor, involving E_0 in Equation (1). Then we use the same approximation [11, 12] that O'Grady *et al.* [8] have used, that is $\mu^2/kTx^3 \ll 1$ which puts some restrictions on the size of the particles and the temperature of the system, but is nevertheless useful to give an idea of the behavior of the assembly in the dimer model.

2.1. Field Parallel to the Assembly

In this case we assume that the applied magnetic field $\vec{H}$ is in the x-direction as illustrated in Figure 1. The total magnetic energy is given by

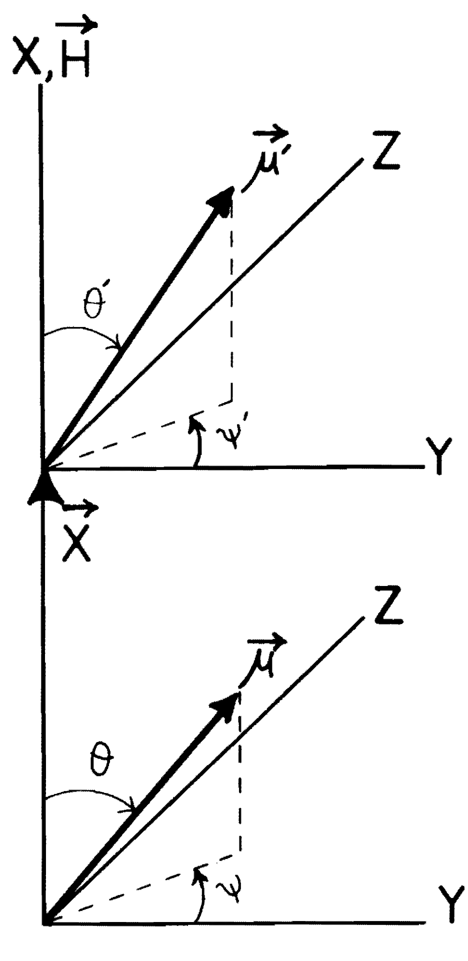

Figure 1.

$$E = \frac{-\mu^2}{x^3} g(\theta, \theta', \psi, \psi') - \mu H(\cos\theta + \cos\theta')$$

where

$$g(\theta, \theta', \psi, \psi') = 2\cos\theta\cos\theta' - \sin\theta\sin\theta'\cos(\psi - \psi').$$

Using the same approximation mentioned earlier, the partition function Z can now be written as the sum of three terms

$$Z = Z_1 + Z_2 + Z_3 \tag{2}$$

where

$$\begin{aligned}
Z_1 &= \int_{x_1}^{x_0} dx \int_0^{\pi} \exp[\alpha\cos\theta]\sin\theta d\theta \int_0^{\pi} \exp[\alpha\cos\theta']\sin\theta' d\theta' \int_0^{2\pi}\int_0^{2\pi} d\psi d\psi', \\
Z_2 &= \frac{\mu^2}{kT}\int_{x_i}^{x_0}\frac{dx}{x^3}\int_0^{\pi}\int_0^{\pi}\int_0^{2\pi}\int_0^{2\pi} g(\theta, \theta', \psi, \psi')\exp[\alpha(\cos\theta + \cos\theta')]\sin\theta\sin\theta' d\theta d\theta' d\psi d\psi' \\
Z_3 &= \frac{\mu^4}{2k^2T^2}\int_{x_i}^{x_0}\frac{dx}{x^6}\int_0^{\pi}\int_0^{\pi}\int_0^{2\pi}\int_0^{2\pi} g(\theta, \theta', \psi, \psi')]^2\exp[\alpha(\cos\theta + \cos\theta')]\sin\theta\sin\theta' d\theta d\theta' d\psi d\psi'
\end{aligned}$$

where

$$\alpha = \frac{\mu H}{kT}. \tag{3}$$

Carrying out the above integrations we find

$$Z_1 = 16\pi^2 (x_0 - x_i)\left(\frac{\sinh\alpha}{\alpha}\right)^2 \tag{4}$$

$$Z_2 = \left[\frac{16\pi^2\mu^2}{kT}\right]\left[\frac{1}{x_i^2} - \frac{1}{x_0^2}\right]\left[\frac{\cosh\alpha}{\alpha} - \frac{\sinh\alpha}{\alpha^2}\right]^2 \tag{5}$$

$$Z_3 = \frac{16\pi^2\mu^4}{5k^2T^2}\left[\frac{1}{x_i^5} - \frac{1}{x_0^5}\right]\left[2\left\{\frac{\sinh\alpha}{\alpha} - \frac{2}{\alpha}\left(\frac{\cosh\alpha}{\alpha} - \frac{\sinh\alpha}{\alpha^2}\right)\right\}^2 + \frac{1}{\alpha^2}\left(\frac{\cosh\alpha}{\alpha} - \frac{\sinh\alpha}{\alpha^2}\right)^2\right]. \tag{6}$$

Equations (4), (5), and (6) can be written in terms of the modified spherical Bessel functions i_0 and i_1 [13]

$$\begin{aligned}
Z_1 &= 16\pi^2 (x_0 - x_i)[i_0(\alpha)]^2 \\
Z_2 &= \frac{16\pi^2\mu^2}{kT}\left[\frac{1}{x_i^2} - \frac{1}{x_0^2}\right][i_1(\alpha)]^2 \\
Z_3 &= \frac{16\pi^2\mu^4}{5k^2T^2}\left[\frac{1}{x_i^5} - \frac{1}{x_0^5}\right] f(\alpha)
\end{aligned}$$

where

$$i_0(\alpha) = \frac{\sinh\alpha}{\alpha}, \quad i_1(\alpha) = \frac{\cosh\alpha}{\alpha} - \frac{\sinh\alpha}{\alpha^2},$$

$$f(\alpha) = 2i_0^2(\alpha) - \frac{8}{\alpha}\, i_1(\alpha) i_0(\alpha) + \frac{9}{\alpha^2}\, i_1^2(\alpha).$$

We can write Z of Equation (2) as

$$Z = C_1\, i_0^2(\alpha) + \frac{C_2}{T}\, i_1^2(\alpha) + \frac{C_3}{T^2}\, f(\alpha) \tag{7}$$

with

$$C_1 = 16\pi^2\,(x_0 - x_i)\,,$$

$$C_2 = \frac{16\pi^2\mu^2}{k}\left[\frac{1}{x_i^2} - \frac{1}{x_0^2}\right]$$

and

$$C_3 = \frac{16\pi^2\mu^4}{5k^2}\left[\frac{1}{x_i^5} - \frac{1}{x_0^5}\right].$$

The magnetization M for a system consisting of N non-interacting identical particles is given by

$$M = kT\frac{\partial \ln Z_N}{\partial H}$$

where Z_N is the N-particle partition function. By analogy, M for $N/2$ non-interacting pairs is given by

$$M = kT\frac{\partial \ln Z_N/2}{\partial H} \tag{8}$$

with $Z_N/2 = (Z)^{N/2}$. Equation (8) becomes

$$M = \frac{NkT}{2Z}\frac{\partial Z}{\partial H}. \tag{9}$$

Differentiating Equation (7) with respect to H and substituting into Equation (9) we get

$$M = N\mu\frac{C_1 i_0(\alpha)\, i_1(\alpha) + \frac{C_2}{T}\, i_1(\alpha)\left[i_0(\alpha) - \frac{2i_1(\alpha)}{\alpha}\right] + \frac{C_3}{T^2}G(\alpha)}{C_1 i_0^2(\alpha) + \frac{C_2}{T}\, i_1^2(\alpha) + \frac{C_3}{T^2}\, f(\alpha)} \tag{10}$$

where

$$G(\alpha) = 2i_0(\alpha)i_1(\alpha) - \frac{4}{\alpha}\left[i_1^2(\alpha) + i_0^2(\alpha)\right] + \frac{21}{\alpha^2}\, i_1(\alpha)i_0(\alpha) - \frac{27}{\alpha^3}\, i_1^2(\alpha). \tag{11}$$

As a check on the validity of the above result we should be able to retrieve the Langevin function by assuming that our system is non-interacting. In Equation (10) we simply set $C_2 = C_3 = 0$ and we get

$$M = N\mu \frac{i_1(\alpha)}{i_0(\alpha)} = N\mu L(\alpha). \tag{12}$$

In order to calculate M for low fields and high temperatures we assume $\alpha \ll 1$. Using $i_0(\alpha) \approx 1 + \alpha^2/6$ and $i_1(\alpha) \approx \alpha/3(1 + \alpha^2/10)$ we find that $f(\alpha) \approx 1/3$ and $G(\alpha) \approx 7\alpha/45$. Hence M can be written as

$$M = \frac{N\mu\alpha}{3} \frac{C_1 + \dfrac{C_2}{3T} + \dfrac{7C_3}{15T^2}}{C_1 + \dfrac{C_3}{3T^2}}. \tag{13}$$

Now, the initial susceptibility X_i is defined as

$$\begin{aligned} X_i &= \left(\frac{\partial M}{\partial H}\right)_{H\to 0} \\ &= \frac{N\mu^2}{3kT} \frac{1 + \dfrac{C_2}{3C_1T} + \dfrac{7C_3}{15C_1T^2}}{1 + \dfrac{C_3}{C_1T^2}} \end{aligned} \tag{14}$$

which can be written using the Taylor series expansion and neglecting terms of order higher than $1/T$ as

$$X_i = \frac{N\mu^2}{3k} \frac{1}{T - \dfrac{C_2}{3C_1}}. \tag{15}$$

So, X_i behaves like Curie–Weiss law with Curie temperature

$$T_0 = \frac{C_2}{3C_1} = \frac{\mu^2}{3k} \frac{x_i + x_0}{x_0^2 x_i^2}. \tag{16}$$

2.2. Field Perpendicular to the Assembly

In this case we choose the magnetic field in the z direction and perpendicular to the assembly which is taken along the x direction as shown in Figure 2. The interaction energy becomes

$$E_0 = \frac{\mu^2}{x^3} g_1(\theta, \theta'\psi, \psi').$$

where

$$g_1(\theta, \theta', \psi, \psi') = \cos\theta\cos\theta' + \sin\theta\sin\theta'\cos(\psi - \psi') - 3\sin\theta\sin\theta'\cos\psi\cos\psi',$$

and the total energy becomes

$$E = E_0 - \mu H(\cos\theta + \cos\theta').$$

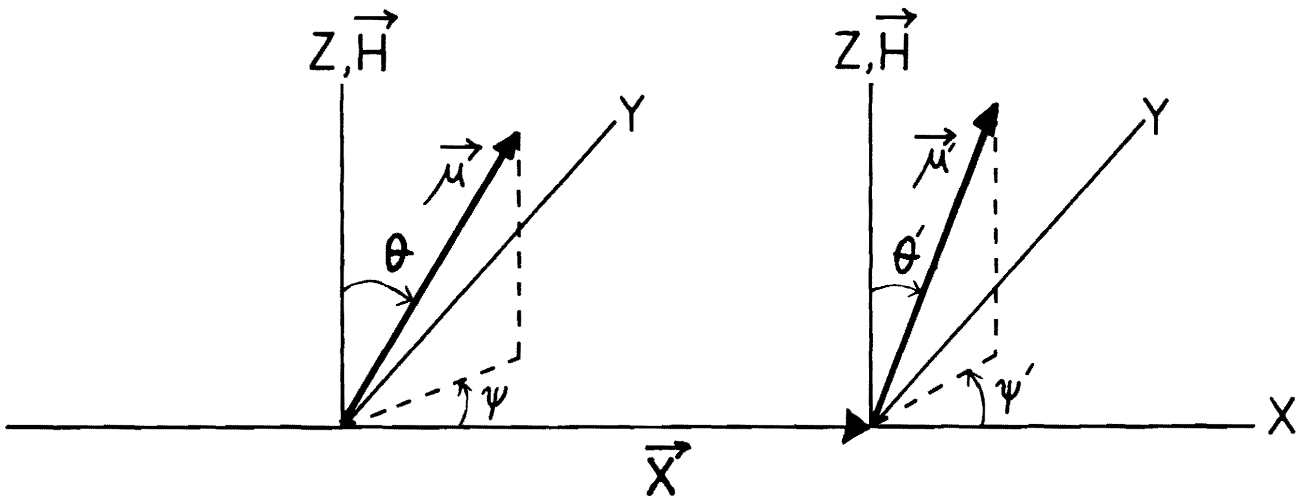

Figure 2.

Now to calculate the partition function, we use the same method and approximation that has been used in Section (2.1), so we write the partition function Z as the sum of Z_1, Z_2, and Z_3. After carrying out the necessary integrations, we find

$$\begin{aligned} Z_1 &= 16\pi^2 (x_0 - x_i)\, i_0^2(\alpha) \\ Z_2 &= -\frac{8\mu^2\pi^2}{kT}\left(\frac{1}{x_i^2} - \frac{1}{x_0^2}\right) i_1^2(\alpha) \\ Z_3 &= \frac{8\mu^4\pi^2}{5k^2T^2}\left[\frac{1}{x_i^5} - \frac{1}{x_0^5}\right]\left[i_0^2(\alpha) - \frac{4}{\alpha}\, i_0(\alpha) i_1(\alpha) + \frac{9}{\alpha^2}\, i_1^2(\alpha)\right]. \end{aligned}$$

The partition function Z can now be written as

$$Z = C_1' i_0^2(\alpha) + \frac{C_2'}{T}\, i_1^2(\alpha) + \frac{C_3'}{T^2}\, f_1(\alpha)$$

where

$$\begin{aligned} C_1' &= 16\pi^2 (x_0 - x_i) = C_1 \\ C_2' &= \frac{8\mu^2\pi^2}{k}\left[\frac{1}{x_i^2} - \frac{1}{x_0^2}\right] = -\frac{C_2}{2}, \\ C_3' &= \frac{16\mu^4\pi^2}{5k^2}\left[\frac{1}{x_i^5} - \frac{1}{x_0^5}\right] = C_3 \end{aligned}$$

and

$$f_1(\alpha) = \frac{1}{2}\left[i_0^2(\alpha) - \frac{4}{\alpha}\, i_0(\alpha) i_1(\alpha) + \frac{9}{\alpha^2}\, i_1^2(\alpha)\right].$$

Now, using Equation (9) we find that

$$M = N\mu \frac{C_1' i_0 i_1 + \dfrac{C_2'}{T}\, i_1 \left(i_0 - \dfrac{2i_1}{\alpha}\right) + \dfrac{C_3'}{T^2}\, G_1(\alpha)}{C_1' i_0^2 + \dfrac{C_2'}{T}\, i_1^2 + \dfrac{C_3'}{T^2}\, f_1(\alpha)} \tag{17}$$

where

$$G_1(\alpha) = i_0 i_1 - \frac{2}{\alpha}\left(i_0^2 + i_1^2\right) + \frac{15}{\alpha^2}\, i_0 i_1 - \frac{27}{\alpha^3}\, i_1^2.$$

In the absence of interactions ($C_2' = C_3' = 0$) we obtain

$$M = N\mu L(\alpha) \tag{18}$$

as expected.

In the case of high temperature and low field $\alpha \ll 1$, we find

$$M = \frac{N\mu\alpha}{3} \frac{C_1' + \dfrac{C_2'}{3T} + \dfrac{8C_3'}{15T^2}}{C_1' + \dfrac{C_3'}{T^2}} \tag{19}$$

and

$$X_i = \frac{N\mu^2}{3kT} \frac{C_1' + \dfrac{C_2'}{3T} + \dfrac{8C_3'}{15T^2}}{C_1' + \dfrac{C_3'}{T_2}}. \tag{20}$$

Using the same approximation discussed before we obtain

$$X_i = \frac{N\mu^2}{3k} \frac{1}{T - \dfrac{C_2'}{3C_1'}} \tag{21}$$

which is the Curie–Weiss law with Curie temperature

$$T_0 = \frac{C_2'}{3C_1'} = -\frac{\mu^2}{6k} \frac{(x_i + x_0)}{x_i^2 x_0^2}. \tag{22}$$

In magnitude this is one half the Curie temperature obtained in Section (2.1) and is negative.

3. DISCUSSION AND CONCLUSION

We have seen that, using the dimer model in a one-dimensional assembly of dilute ferrofluid at high temperature and low applied magnetic field, the initial susceptibility follows the Curie–Weiss law both in cases of field parallel and perpendicular to the system. However, in magnitude, the Curie temperature in the case of the field parallel to the system is twice that when the field is perpendicular to the system. Moreover the Curie temperature is negative in the second case. The appearance of the Curie temperature, in contrast to the model predicted by Chantrell *et al.* [10], is due to the interparticle dipole–dipole interactions. These interactions tend to increase the magnetization, and consequently the initial susceptibility of the system when the magnetic field is parallel to the linear assembly but they tend to decrease the magnetization, and consequently the initial susceptibility when the magnetic field is perpendicular to the assembly.

Finally we point out that this theory may be tested by taking a ferrofluid textured in the form of chain aggregates in the presence of an applied magnetic field. If the field is turned off the chain structure (texture) persists for a certain relaxation time [12]. If a magnetic field is now applied and the initial susceptibility is promptly measured as a function of temperature, we predict that the Curie temperature will be given reasonably well by the expressions (16) and (22) for the cases of field parallel and perpendicular to chains, respectively.

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