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The aggregation mechamism of small magnetic particle chains

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The aggregation mechanism of small ferromagnetic particle chains in viscous media is discussed. If we describe such chains as infinite uniformly magnetized cylinders, there will be no interchain interaction. It is shown that in the next approximation – chains of identical (i.e. spherical) particles – the interaction force decreases exponentially with the interchain distance ($f \sim \exp(-r/a)$, where a is the particle diameter), and hence this model cannot explain the strong aggregation of chains. However, the distribution of particle magnetic moments (resulting from size, shape etc. distributions) leads to an interaction, decreasing only as $f \sim r^{-6}$ which can probably explain the observed phenomena.

It is well known that for a ferromagnetic particle assembly in viscous media strong aggregation occurs due to the interparticle magnetostatic interaction. If the system is placed in a sufficiently large external field (i.e. during the orientation procedure for magnetic tapes and disks), then firstly long chains are formed, and thereafter these chains aggregate into large filament-like clusters. The process of single-chain formation was described theoretically by de Gennes [1] and Jordan [2] and intensively studied experimentally (see references in ref. [3]). But the mechanism of the chain aggregation was not understood until recently [4,5]. In the continuous approximation, when we consider straight chains as thin cylinders, the interchain interaction does not exist, because the infinite uniformly magnetized cylinder does not induce any magnetic field in the surrounding space. The first attempt [5] to explain the interchain interaction takes the discrete chain structure into account. For the simplest case of spherical particles of diameter d this model describes the interaction of two straight infinite chains of dipoles with a distance d between the dipoles within the chain. The interaction force f per unit length is $f = F_{1p}/d$, where F_{1p} denotes the force acting on one particle.

For a particle at a distance a from the infinite chain this force can be, in general, found from the usual expression for the dipole interaction, which results in a power series. But this series cannot be summarized analytically and due to its slow convergence attempts to solve it numerically can lead to quantitatively incorrect results, and even to the wrong sign of the interchain interaction [5].

To solve this model we shall use the potential theory methods. The magnetostatic force acting on a given particle with magnetic moment m from all the particles of the other chain can be found as

$$F_{1p} = -\nabla(\boldsymbol{m} \cdot \boldsymbol{H}) = -\nabla(\boldsymbol{m} \cdot \nabla\phi),$$

where ϕ is the scalar magnetic potential of the chain, which can be found from the Poisson equation $\nabla\phi = -4\pi\rho$. For the chain of dipoles the (magnetic) charge density ρ is the sum of δ-function derivatives [6]

$$\rho(a, z) = m\delta(a)\sum_n \delta'(z - nd). \quad (1)$$

Due to the cylindrical symmetry the transition to the Fourier components in the Poisson equation results in

$$\phi(k_\perp, k_z) = -4\pi Z(kz)R(k_\perp)/(k_\perp^2 + k_z^2),$$

where $Z(k_z)$ and $R(k_\perp)$ are the Fourier components of the charge density written in the form $\rho(a, z) = R(a)Z(z)$. For the chain with the charge density (1) $R(k_\perp) = 1$ and

$$Z(k_z) = \text{im } k_z\sum\delta(k_z - 2\pi l/d), \quad l = 1,2,\ldots.$$

Returning to the space coordinates we obtain

$$\phi(a, z) = 8\pi\sum_l k_l K_0(k_l a)\sin(k_l z),$$

where $k_l = 2\pi l/d$ and K_0 denotes the modified Bessel function [7]. Using the asymptotic expression of $K_0(x)$ for $x \gg 1$ ($a \gg k_1^{-1} \sim d$), we obtain the desired expression for ϕ in the limit of most interest $a \gg d$ with the largest contribution from the first term ($l = 1$) decaying as $\phi(a) \sim e^{-2\pi a/d}$. Hence the potential of the infinite chain of dipoles decays exponentially with the characteristic radius $r_c = d/2\pi$, which is much less than the particle diameter. It is obvious that the interchain interaction force will follow the same exponential decay law and therefore the model of infinite dipole chains cannot explain the observed strong chain aggregation.

We propose an extension of this model, which takes into account the distribution of particle sizes, shapes etc. resulting in the distribution of particle magnetic moments. In the continuous approximation this leads to fluctuations of the chain magnetic moment density around its average value. Such a chain produces in the

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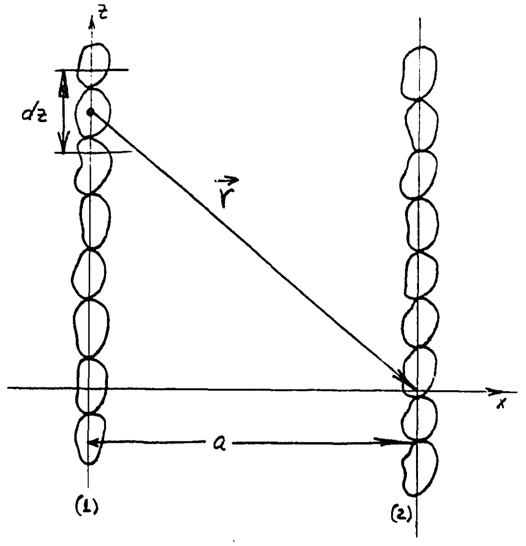

Fig. 1. The coordinate system for two interacting chains.

surrounding space a random magnetic field. This field acts on the moments of other chains, leading to an (average) attractive interaction, in analogy with the self-consistent fluctuations of the electric dipole moment leading to the van der Waal's attractive interaction between neutral atoms [8].

To solve the problem exactly, consider two parallel chains with an interchain distance a (fig. 1). We choose the z-axis of our coordinate system along the first chain, and the x-axis will be aligned to the second chain. The magnetic moment density of the first chain is $\mu(z) = \mu_{av} + \Delta\mu(z)$, with small fluctuations: $\langle \Delta\mu^2 \rangle^{1/2} \ll \mu_{av}$. Then the total force, acting on the particle with magnetic moment $\boldsymbol{m} = m\boldsymbol{e}_m$ from the second chain, is

$$\boldsymbol{F}_{1p} = -\nabla U = \nabla(\boldsymbol{m} \cdot \boldsymbol{H}_1) = m\nabla(\boldsymbol{e}_m \cdot \boldsymbol{H}_1),$$

where $\boldsymbol{H}_1$ denotes the field from the first chain at the location point of the moment $\boldsymbol{m}$. Due to the symmetry of the problem, after averaging over different $\boldsymbol{H}_1$-configurations only the x-component of the force $F_{x,1p}$ is non zero:

$$F_{x,1p} = m\frac{\partial}{\partial x}\left(\langle e_{mx}H_{1x}\rangle + \langle e_{my}H_{1y}\rangle + \langle e_{mz}H_{1z}\rangle\right),$$

where the second term vanishes due to the same symmetry. For small fluctuations the angle α between the moments and the z-axis is small and to linear accuracy $e_z \approx 1 - \alpha^2/2 \approx 1$, which results in

$$\langle e_{mz}H_{1z}\rangle \approx \langle H_{1z}\rangle = 0$$

And hence

$$F_{x,1p} = m\frac{\partial}{\partial x}\left(\langle e_{mx}H_{1x}\rangle\right). \tag{2}$$

From the equilibrium equation for the magnetic moment of the particle with uniaxial anisotropy in an external field one can easily obtain that for the weak field (the case due to weak moment fluctuations) $e_{mx} = 0.5H_x/\beta I_s$ [9], where β denotes the reduced anisotropy constant [10] (for a cylinder $\beta = 2\pi$) and I_s is the saturation magnetization. Substituting this expression into (2) and transforming the derivative variable to a, we obtain

$$F_{x,1p} = \frac{m}{2\beta I_s}\frac{\partial}{\partial a}\left(\langle H_{1x}(a)^2\rangle\right). \tag{3}$$

Let us define the dispersion of the x-component of the random field. The element $\mathrm{d}z$ of the chain with the coordinate z and dipole moment $\mu(z)\,\mathrm{d}z$ produces at the location point of the given particle (see fig. 1) the field $\mathrm{d}\boldsymbol{H}_1 = (3(\boldsymbol{e}_z\boldsymbol{e}_r)\boldsymbol{e}_r - \boldsymbol{e}_z)\mu(z)/r^3$, where $\boldsymbol{e}_r = \boldsymbol{r}/r$ with the x-component $\mathrm{d}H_{1,x} = 3az\mu(z)/r^5$, and the dispersion is

$$\langle H_{1,x}^2(a)\rangle = 9a^2\int\int \mathrm{d}z\,\mathrm{d}z'\frac{zz'\langle\Delta\mu(z)\Delta\mu(z')\rangle}{\left[(a^2+z^2)(a^2+z'^2)\right]^{5/2}}.$$

We are interested in the field at the distance $a \gg d$ and hence the correlator in (4) can be considered as a δ-function:

$$\langle\Delta\mu(z)\Delta\mu(z')\rangle = M\delta(z-z').$$

Then the integral (4) can be evaluated and the force per unit length is

$$f_x = -\frac{225}{512}\frac{VM}{d}\frac{1}{a^6}, \tag{5}$$

where the measure of the moment fluctuations M can be written using the dispersion of the particle magnetic moment as

$$M = \langle\Delta m^2\rangle/d = \langle m\rangle^2\sigma^2/d$$

with the average moment $\langle m\rangle$ and the reduced dispersion $\sigma^2 = \langle\Delta m^2\rangle/\langle m\rangle^2$. Substituting this expression into (5), we obtain the final expression, which is useful in practice, for the interaction force per unit length between two chains

$$f_x = -C/a^6,$$

where

$$C = \frac{225}{512}\frac{\langle m\rangle^2\sigma^2 V}{d^2}.$$

We underline two features of this result. First of all, this is the attractive interaction (negative x-component of the force, see fig. 1). Secondly, this force decays as a^{-6}, not exponentially. This means that the interaction obtained has a long-range character, in contrast with the model considered in the first part of this paper. The constant C, which determines the absolute value of the interaction force, is proportional to the dispersion of the particle moments σ^2, and hence the interaction is weaker for more homogeneous chains.

In conclusion, we note that the proposed model allows us to explain the strong aggregation of chains consisting of small ferromagnetic particles. Further investigations must be performed to study the aggregation kinetics of such chains.

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