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Assembly of magnetic spheres in strong homogeneous magnetic field

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HIGHLIGHTS

- Phase diagram of ground states for finite number of magnetic beads in strong external field.
- Two-chain state characterized by tails of constant size.
- Relevance of the key interaction between a chain and a single bead.

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ABSTRACT

The assembly in two dimensions of spherical magnets in strong magnetic field is addressed theoretically. It is shown that the attraction and assembly of parallel magnetic chains is the result of a delicate interplay of dipole–dipole interactions and short ranged excluded volume correlations. Minimal energy structures are obtained by numerical optimization procedure as well as analytical considerations. For a small number of constitutive magnets $N_{\mathrm{tot}} \leq 26$, a straight chain is found to be the ground state. In the regime of larger $N_{\mathrm{tot}} \geq 27$, the magnets form two touching chains with equally long tails at both ends. We succeed to identify the transition from two to three touching chains at $N_{\mathrm{tot}} = 129$. Overall, this study sheds light on the mechanisms of the recently experimentally observed ribbon formation of superparamagnetic colloids via lateral aggregation of magnetic chains in magnetic field (Darras et al., 2016).

1. Introduction

There are several reasons for strong and growing interest in self-assembled structures of dipolar particles (i.e., with electric or magnetic dipoles). On the technological side, such systems have enormous potential applications. For instance, manufacturing of novel optical and stimuli-responsive materials is based on self assembly of magnetic particles [1,2]. On the other hand, dipolar particles and the resulting phases can be well tuned by imposing an external field [3,4]. Assemblies of magnetic particles are known to produce a plethora of one-, two-, and three dimensional objects (e.g., chains, rings, and even tubes) [5–7]. Experimental evidence of spontaneous assembly of small magnetic nanocubes into highly ordered chains, sheets, and cuboids in solution by applying a magnetic field was recently reported [8]. Equally intriguing microstructures such as Saturn ring- or flower-like structures were also found experimentally for a mixture of paramagnetic and diamagnetic colloidal particles within a magnetized ferrofluid [9]. From the perspective of biophysics, magnetic particles can be regarded

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as a model system for probing the polar organization of microtubules [10] or generating spontaneous helical superstructures [11,12] reminiscent of DNA molecules.

By essence, the dipole–dipole driving force for self-assembly is long range and highly anisotropic (i.e., non-central pair potential) [13,14] and therefore represents a formidable theoretical challenge. In this spirit, the pioneering theoretical work of Jacobs and Beans [15] and later that of de Gennes and Pincus [16] about the microstructure of self-assembled (spherical) magnets shed some light on the ordering mechanisms. More recently, microstructures of dipolar fluids have been thoroughly studied by computer simulations [17–19] and in experiments [20]. There, an important common feature is the formation of chains [17,19] and possibly in presence of an external magnetic field [18,20]. The interaction of two *infinite* chains in strong external magnetic field was studied analytically in the early 90's [21,22]. Depending on their relative shift, short-range attractions/repulsion sets in with a roughly exponential decay. With all that being said, only recently, the ground state structures of magnetic spheres without external magnetic field have been properly addressed in three dimensions [6,23,24] as well as in two dimensions [7].

The goal of the present contribution is to tackle the fascinating problem of self-assembly of magnets under strong magnetic field in a physically simple and transparent framework. Motivated by the self-assembly experimentally revealed with a small number of magnetic beads [8,9,25,26], we explore (effective) interactions and assemblies of a finite number of magnetic beads with *parallel dipoles* (e.g., as obtained by a strong external magnetic field) and confined in two dimensions (e.g., by gravity [26] and/or by capillarity at liquid/liquid interface [25]).[1] When dealing with the search of the ground state, we utilize two fully different routes to calculate the energy minimum of the system: (i) genetic algorithm and (ii) direct calculation and comparison of the energy of different configurations. The paper is organized as follows: In Section 2 we expose the magnetic chains Hamiltonian. Section 3 is devoted to the analytical results dealing with the two-chain state. Phase diagram of self-assembled magnets obtained by numerical genetic algorithm is discussed in Section 4. Concluding remarks are provided in Section 5.

2. Model

2.1. Pair interaction potential

We begin by considering two magnetic hard spheres of diameter d separated by a distance $r_{12} = |\vec{r}_2 - \vec{r}_1| > d$, where $\vec{r}_1$ and $\vec{r}_2$ represent the position vectors of the centers of particle 1 and particle 2, respectively. These spherical magnets being also characterized by a magnetic moment $(\vec{m}_1, \vec{m}_2)$, the pair potential energy is dictated by:

$$U(\vec{r}_{12}) = C\frac{1}{r_{12}^3}\left[\vec{m}_1 \cdot \vec{m}_2 - 3\frac{(\vec{m}_1 \cdot \vec{r}_{12})(\vec{m}_2 \cdot \vec{r}_{12})}{r_{12}^2}\right], \tag{1}$$

where C is a constant that depends on the intervening medium (e.g., for vacuum $C = \frac{\mu_0}{4\pi}$ with μ_0 being the vacuum permeability).

The approach we adopt in this work is based on the calculation of the magnetic energy of various configurations of spheres in externally imposed magnetic field $\vec{B} = B\vec{e}_x$ aligned with x-axis. We assume that the external magnetic field is strong, i.e, $B \gg mC\frac{1}{d^3}$ (with $m := |\vec{m}_1| = |\vec{m}_2|$), so that all dipole moments in the system are aligned with $\vec{B}$ and hence parallel to the x-axis, i.e. $\vec{m} = m\vec{e}_x$. In this limit of strong field, the pair potential (1) becomes

$$U(\vec{r}_{12}) = C\frac{m^2}{r_{12}^3}\left[1 - 3\frac{(x_2 - x_1)^2}{r_{12}^2}\right]. \tag{2}$$

2.2. Chains Hamiltonian

It is convenient to introduce the energy scale defined by $U_{\uparrow\uparrow} \equiv \frac{Cm^2}{d^3}$ that physically represents the repulsive potential value for two parallel dipoles at contact standing side by side as clearly suggested by the notation. The dipoles attract if placed with head-to-tail (i.e., $\rightarrow\rightarrow$). The latter promotes formation of one dimensional chains consisting of many dipoles (i.e., $\rightarrow\rightarrow \ldots \rightarrow$). The reduced total potential energy of interaction of a system consisting of two chains made up of N_1 and N_2 particles, $U^{tot}_{N_1N_2}$, can be written as

$$U^{\mathrm{tot}}_{N_1N_2} = \frac{1}{2}\sum_{\substack{i,j=1\\ i\neq j}}^{N_1+N_2}\frac{U(\vec{r}_{ij})}{U_{\uparrow\uparrow}} \quad (r_{ij} \geq d). \tag{3}$$

[1] Note that our model corresponds to a zero temperature approach where fluctuations (for instance in chain length and/or shape) [27] are neglected. Hence, when comparing theory vs experiments, one has to bear in mind that only the high magnetic coupling (external field) is relevant. This limit is typically reached in magnetorheological fluids or superparamagnetic colloids with saturated magnetization under the influence of a strong external field.