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On minimal energy dipole moment distributions in regular polygonal agglomerates

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ABSTRACT

Static, regular polygonal and close-packed clusters of spherical magnetic particles and their energy-minimizing magnetic moments are investigated in a two-dimensional setting. This study focuses on a simple particle system which is solely described by the dipole-dipole interaction energy, both without and in the presence of an in-plane magnetic field. For a regular polygonal structure of n sides with $n \geq 3$, and in the absence of an external field, it is proved rigorously that the magnetic moments given by the roots of unity, i.e. tangential to the polygon, are a minimizer of the dipole–dipole interaction energy. Also, for zero external field, new multiple local minima are discovered for the regular polygonal agglomerates. The number of found local extrema is proportional to $\lfloor n/2 \rfloor$ and these critical points are characterized by the presence of a pair of magnetic moments with a large deviation from the tangential configuration and whose particles are at least three diameters apart. The changes induced by an in-plane external magnetic field on the minimal energy, tangential configurations are investigated numerically. The two critical fields, which correspond to a crossover with the linear chain minimal energy and with the break-up of the agglomerate, respectively are examined in detail. In particular, the numerical results are compared directly with the asymptotic formulas of Danilov et al. (2012) [23] and a remarkable agreement is found even for moderate to large fields. Finally, three examples of close-packed structures are investigated: a triangle, a centered hexagon, and a 19-particle close packed cluster. The numerical study reveals novel, illuminating characteristics of these compact clusters often seen in ferrofluids. The centered hexagon is energetically favorable to the regular hexagon and the minimal energy for the larger 19-particle cluster is even lower than that of the close packed hexagon. In addition, this larger close packed agglomerate has two distinctive regimes in the magnetization, which corresponds to two very different susceptibilities, in marked contrast to the behavior observed in regular polygonal structures.

1. Introduction

Ferrofluids are a class of magnetic fluids consisting of a suspension of magnetic particles with diameter of about 10 nm in a Newtonian liquid. Stable ferrofluids were developed in the 1960 s and have since been the subject of substantial investigation [1,2]. Ferrofluids have an enormous potential for applications due to their strong response to an external magnetic field while retaining their fluidity. In recent years, several ferrofluid applications were developed in industry and medicine [3–6]. However, the intricate magnetic field-particle-flow interaction still poses many challenges for achieving a better understanding, control, and predictability of this important class of complex fluids.

The first analytical models used to investigate ferrofluids did not take in account the dipole–dipole interactions among particles, treating the suspension as an isotropic dilute gas [7–9] but it was soon recognized that these interactions are important. Moreover, the suspended magnetic particles form agglomerates, specially chains and clusters in the presence of even moderate magnetic fields, and these structures significantly affect the macroscopic behavior of the ferrofluid [10–13]. In the two-dimensional setting of a thin film it has been found, both experimentally and numerically, that the suspended magnetic particles group together in complicated aggregates, as chain, small rings, and close packed structures, with or without an external magnetic field [14–20]. In particular, the small rings are reported to be very stable unless subjected to a sufficiently strong field.

Guo et al. [21] performed experiments with no applied field using magnetic balls of 20 μm in diameter by randomly perturbing them with a vibrational table. They found that the probability of

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forming a ring with n particles is a Poisson-like distribution with a peak at $n=9$. They also obtained, via molecular dynamics, the critical *in-plane* field to break the ring as a function of n and found this relation to be also Poisson-like with a peak at $n=9$. They concluded that the 9-particle ring is the most stable one.

Jund et al. [22] did a study of these ring-shaped structures using molecular dynamics simulations. They showed that an initial polygonal aggregate will break abruptly with an increasing magnetic field in the direction *perpendicular* to the polygon plane. Such a non-smooth topological transition has also been demonstrated numerically by Danilov et al. [23], who showed that the ring structure changes to a beautiful, centered hexagonal spaced configuration as the applied field is increased.

For an *in-plane* applied field, the ring-shaped agglomerates break and form chains aligned in the field direction. Morimoto et al. [24], using Monte Carlo simulations, found that a field increase results in a reduction of the probability of finding a ring. Prokopieva et al. [25] identified, also via Monte Carlo simulations, a jump in magnetization with increasing magnetic field, indicating the rupture of the ring and its transformation into a chain.

In both perpendicular and in-plane field settings, it has been observed that the agglomerates with initial polygonal shape keep this configuration (with slight modifications) up until very close to the structural break-up. Kun et al. [26] presented evidence of this for aggregates of much bigger magnetic particles, with diameter of about 50 μm. These particles are used in the composition of advanced magnetorheological suspensions, another type of magnetic fluid. They observed that the *rate of increase* of the magnetic field will affect the after break-up chain configuration. The effect of the Brownian motion in particles of this size ($\approx$50 μm) is negligible compared to the dipolar interactions.

Our focus in this work is to study analytically and numerically minimal energy, magnetic moment configurations (ground states) of *planar*, *static* polygonal and close packed aggregates of monodisperse magnetic spheres. To keep our setting two-dimensional, we only consider the cases of zero field and when there is an applied *in-plane* field. Moreover, we assume that the system is dominated by the dipole-dipole interaction and both thermal and steric effects are neglected.

First, for a regular polygonal structure with n in-contact particles, $n \geq 3$, and zero applied field, Prokopieva et al. [27] concluded, by splitting the energy in pair interactions, that the magnetic moments oriented tangential to the polygon constitute an energy minimizer. We provide an independent, rigorous proof of this result (in the Apendix) and present novel numerical results that show the existence of multiple local minima for the regular polygonal agglomerates. The number of the numerically found local extrema is proportional to $\lfloor n/2 \rfloor$ (the integer part of $n/2$, for $n > 7$) and these local minima are characterized by the presence of a pair of magnetic moments with a large deviation from the tangential configuration and whose particles are at least three diameters apart.

We investigate too the effect of an in-plane external magnetic field on the minimal energy, tangential configurations with Monte Carlo simulations. We examine in detail the two critical fields, which correspond to a crossover with the linear chain minimal energy and with the break-up of the agglomerate, as well as the overall magnetization of the structure. Danilov et al. [23] have obtained corresponding asymptotic, analytic results by considering a small angle perturbation from the tangential configuration. However, their numerical setting at finite, nonzero temperature did not allow for a full exploration of these asymptotic results. Our simpler static, planar, and zero temperature framework permits us to do this in great detail. We find that the asymptotic formulas of Danilov et al. [23] offer a remarkably accurate prediction of the break-up critical field, the magnetization, and susceptibility of the static polygonal agglomerates, even for moderate to large fields.

Finally, we investigate three examples of close-packed structures as illustrative instances of the close-packed clusters often observed in magnetic fluids [14]. We consider a triangle, a centered hexagon, and a 19-particle close packed agglomerate. We obtain the minimal energy and equilibrium magnetization of these compact structures. In particular, we find that the centered-hexagon is energetically favorable to the regular hexagon and that the 19-particle cluster has an even lower minimal energy. Moreover, this larger aggregate has two distinctive regimes in the magnetization, which corresponds to two very different susceptibilities, in marked contrast to the behavior observed in regular polygonal structures.

This article is organized as follows. In Section 2, we give the geometry description and other details of the problem formulation. In Section 3, we look at the regular polygonal agglomerates in the absence of an external field and provide, in the Appendix A, a proof that the n-th roots of unity correspond to a minimum of the energy for $n \geq 3$. Then, in Section 4, we examine numerically the effect of an in-plane magnetic field on the minimal energy, tangential configurations. Section 5 is devoted to the investigation of local minima of the dipole–dipole energy and Section 6 to three illustrative examples of close-packed structures. Finally, further discussion of the physical significance of the results and concluding remarks are presented in Section 7.

2. Problem formulation

We consider a system of n identical, spherical magnetic particles, dominated by the dipole-dipole interaction (thermal, hydrodynamic, and steric effects are neglected). Denoting the center of the particles and their magnetic moments by $\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n$ and $m\hat{\mathbf{u}}_1, m\hat{\mathbf{u}}_2, \ldots, m\hat{\mathbf{u}}_n$, respectively, and in the presence of an external magnetic field $\mathbf{H} = H\hat{\mathbf{h}}$, the system's energy is given by [1]:

$$\Phi_n = \sum_{i<j}^{n} \frac{\mu_o m^2}{4\pi r_{ij}^3}\left[(\hat{\mathbf{u}}_i\cdot\hat{\mathbf{u}}_j) - 3(\hat{\mathbf{u}}_i\cdot\hat{\mathbf{r}}_{ij})(\hat{\mathbf{u}}_j\cdot\hat{\mathbf{r}}_{ij})\right] - \sum_{j=1}^{n} \mu_0 m H \hat{\mathbf{h}}\cdot\hat{\mathbf{u}}_j, \tag{1}$$

where $\mu_o = 4\pi \times 10^{-7}\ \mathrm{N/A^2}$ is the vacuum magnetic permeability, and $\boldsymbol{r}_{ij} = \boldsymbol{r}_j - \boldsymbol{r}_i = r_{ij}\hat{\mathbf{r}}_{ij}$. The caret denotes unit vectors. The first sum in (1) is the dipole-dipole interaction energy and the second one is the Zeeman energy.

Using the particle diameter d as the characteristic length scale and $\Phi^c = \mu_o m^2/(4\pi d^3)$ as the energy scale (equal to half the energy of two in-contact particles with collinear magnetic moments) we can write $\boldsymbol{\Phi}_n$ in dimensionless form as

$$\Phi_n = \sum_{i<j}^{n} \frac{1}{r_{ij}^3}\left[(\hat{\mathbf{u}}_i\cdot\hat{\mathbf{u}}_j) - 3(\hat{\mathbf{u}}_i\cdot\hat{\mathbf{r}}_{ij})(\hat{\mathbf{u}}_j\cdot\hat{\mathbf{r}}_{ij})\right] - \alpha \sum_{j=1}^{n} \hat{\mathbf{h}}\cdot\hat{\mathbf{u}}_j, \tag{2}$$

where

$$\alpha = \frac{\mu_o m H}{\Phi^c} \tag{3}$$

represents a measure of the strength of the external magnetic field relative to the contact energy.

We consider static agglomerates of in-contact particles forming a regular n-side polygon (an n-gon) as illustrated schematically in Fig. 1. Moreover, we restrict our study to the two-dimensional case in which all the magnetic moments, the center of mass of the particles, and the external field (oriented in the y direction) are constrained to the plane of the polygon. We focus on the problem of finding the orientation (polar) angle of the magnetic moments, $\theta_1, \theta_2, \ldots, \theta_n$, which minimizes $\boldsymbol{\Phi}_n$.