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MORPHOLOGY, STABILITY, AND MORPHOGENESIS OF DIPOLE AGGREGATES

Dedicated to the 70th birthday of Robert Heller

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Abstract. In this paper, we establish some basic thermodynamic formulas and master systems allowing one to explore analytically and calculate numerically equilibrium configurations and quasi-static evolution of crystalline substances with dipolar interaction. All the master systems are certain extensions of the equations of electro- or magnetostatics. To elucidate and emphasize the "dipolar aspect" of the problem, we introduce and explore the simplest possible system with dipolar interaction which we call the dipole aggregate. The dipole aggregates are able to accumulate the dipolar electric or magnetostatic energy as well as the surface energy but cannot accumulate any kind of the internal bulk energy. Our approach is based on the minimum-of-the-total-energy principle, i.e., we assume upfront that the absolute temperature is fixed and constant throughout the whole system. Our analysis of the equilibrium shapes of the dipole aggregates and of their stability is based on the traditional tools of the Gibbs equilibrium thermodynamics, namely, on the calculation of the first and second energy variations. Our analysis of quasi-static evolution of the shape of the dipole aggregates is based on the non-equilibrium thermodynamics of dissipative heterogeneous systems (in the spirit of the Onsager irreversible thermodynamics with linear fluxes).

Key words: morphology, stability, thermodynamics, electromagnetism.

1. INTRODUCTION

Various heterogeneous systems with dipolar interaction are of paramount theoretical and practical importance in many branches of physics, chemistry, biology, engineering, materials science, etc. For some of them, electromagnetic interactions and their energy play a central role. For instance, in the problems of ferromagnetism, ferroelectricity and superconductivity $[1^{-3}]$, in physics of colloids

and liquid crystals $[^{4-6}]$, in modern approaches to shapes of solid crystals and crystal growth $[^7]$, and in other disciplines. Naturally, thermodynamics plays a significant role in corresponding theories. The reader interested in the state-of-the-art of the thermodynamics of electromagnetic media and its applications in continuum physics, mechanics, and materials science can find a wonderful presentation in $[^8]$.

Despite the remarkable progress, which has been achieved in understanding the thermodynamic foundations of electromagnetic interaction (see, for instance, [⁸], as well as older textbooks and reviews [$^{2,9-12}$]), several aspects of the everyoung thermodynamics are yet to be developed in-depth and in-width. For instance, even the very formula of the energy of dipolar interaction or the basic formula of the static ponderomotive force acting on a dielectric body are both still under hot debates. (E.g., The American Journal of Physics publishes several critical papers of that sort annually.)

From the standpoint of thermodynamics, the latter formula is nothing else but a derivation of the first energy variation of a heterogeneous system with a dipolar (electric) interaction. Under these circumstances, it is not surprising that the problem of the second energy variation of such systems has not been even raised in scientific literature, to the best of the author's knowledge. The problem of the second energy variation is instrumental for the analysis of thermodynamic stability. In this respect, the current situation with the second energy variations for the systems with dipolar interaction is exactly the same as it was in the Gibbs thermodynamics of heterogeneous systems with solid components just a couple of decades ago. Actually, for almost a century, the problem of the second energy variation of solid heterogeneous systems with sharp interfaces was completely ignored. That is why thermodynamics of solids has not demonstrated, until recently, any serious interest in morphological instabilities of interfaces. The development of the theory of the second energy variation of heterogeneous solid systems has led to the prediction of several unknown and unusual morphological instabilities, some of which were observed experimentally later on. Currently, these instabilities attract wide interest and rapid progress is achieved regarding both the theory and the experiment.

We hope that future development of the theory of the second energy variation of heterogeneous systems with dipolar interaction will be helpful in establishing novel instability mechanisms in such systems. In contrast to the interfaces in solids, the purely energetic morphological instability of electrically charged interfaces of liquids can be easily observed with a naked eye. This instability has been analysed by Lord Rayleigh more than a century ago. Since that pioneering study, several morphological instabilities of interfaces in the systems with electric or magnetic interactions have been explored for liquid substances. These studies are based on the properly extended hydrodynamic equations rather than on purely thermodynamic analysis. Because of different reasons, the author believes that it is a must for the study of crystalline objects to develop a direct thermodynamic approach to the stability and quasi-static evolution which is equally applicable to solid and liquid substances. Such an approach should be based on the analysis of the first and second energy variations of the systems with dipole interaction.

The isothermal equilibrium and stability of a vast majority of heterogeneous systems with dipolar interaction can be studied with the help of minimization of the following energy functional:

$$E_{total} = \int_{\Omega} d\Omega \psi_b + \int_{\Xi} d\Xi \psi_s + E_{dip}, \tag{1}$$

where E_{total} is the total energy of the system comprising three ingredients: 1) the (local) bulk free energy $\int_{\Omega} d\Omega \psi_b$ with the density ψ_b per unit volume, 2) the surface energy $\int_{\Xi} d\Xi \psi_s$ with the density ψ_s per unit mass, and 3) the non-local dipolar energy E_{dip} . The spatial integral in (1) is actually a sum of the integrals over smooth spatial subdomains occupied by different phases. The surface integral is actually a sum of the integrals over the external surface of the system and the interfaces separating different phases. Depending on the particular problem in question, the bulk free energy density ψ_b can be the function of 1) the elastic displacement gradients ∇U for solids or mass density ρ for liquids, 2) the dipolar electric **P** or dipolar magnetic M momentum of unit volume for dielectric or magnetic media, 3) the gradient of dipolar magnetic momentum $\nabla \mathbf{M}$ for ferromagnetic media, 4) the director d and its spatial gradient ∇d for liquid crystals. The surface energy density is usually a function of the unit normal N to the corresponding interface Ξ . Hence, there is plenty of practically useful applications which can be explored with the energy functional like (1). In the cited literature, the reader can find many presentations of variational techniques of dealing with the first two integrals (which concern both the first and the second energy variations). The last term, the dipolar energy, is quite universal: it is actually the same for all bulk and surface energy models. So, the author thinks it is worthy to analyse it separately of the other two. It is not to say that the dipolar energy is more essential than the other two ingredients; in many cases the dipolar energy is something secondary. But it is the dipolar energy term, the first variation of which has never been analysed sufficiently and second variation has never even been considered (to the best of the author's knowledge). To that end we propose the simplest thermodynamic system the energy of which does not include any bulk energy at all, and that includes the simplest surface energy term only. We call these systems the dipole aggregates (DA).

In what follows, we present (without lengthy details) some traditional thermodynamic issues like 1) calculation of equilibrium shapes, 2) the conditions of morphological stability, and 3) the master system of slow morphological evolution of DA. We disregard many special details of various DAs which are of primary importance in particular applications of DAs and concentrate on the general consequences of the following features:

1) the existence of a local (short-distance) crystalline order,

2) the possibility of rearrangement of elementary "units" of the substance (i.e., the possibility of the boundary units to break their bonds and, then, to migrate and re-attach themselves to the DA in other places of the boundary),

3) the presence of a significant amount of the energy of the surface bonds,

4) the presence of a significant amount of non-local electro- and magnetostatic energies of dipole interaction,

5) the ability to minimize the total energy due to the rearrangement.

To emphasize the most robust implications of these basic features we deliberately ignore other more specific energy sources mentioned above, say, the exchange and the director-field energies which are at the heart of the current theories of ferromagnetism and of liquid crystals, etc. (We do not see any conceptual obstacles for including different ingredients in the bulk free energy.)

Currently, it is widely believed that thermodynamic concepts are valid for a vast variety of systems ranging over more than 20 decimal orders in space and time. From that point of view, it is never known what are the most useful applications of thermodynamic considerations. As an illustration of this thought, we show some implications of the actual consideration to the objects belonging to the opposite side of the length-scale, namely, to the megascopic objects studied in astrophysical and geophysical fluid dynamics. The (in)stabilities of theses objects might have exactly the same thermodynamic nature as some of the nano-objects.

2. A SIMPLE MODEL OF DIPOLE AGGREGATES

For the sake of brevity, in the following we will mention the DAs with electric interactions only (for the thermodynamic aspects raised in this paper the difference between electric and magnetic interactions is purely semantic).

We deliberately limit ourselves to the simplest model of the substance and ignore all external force fields and all ingredients of the accumulated energy E_{ac} . However, we do consider the electric energy of the constituent dipoles E_{el} and the surface energy E_s accumulated at the external boundary S of the volume Ω occupied by the DA (Fig. 1): $E_{ac} = E_{el} + E_s$.

We assume that each elementary unit (or "atom", or "molecule", or cell of periodicity) of the substance has a fixed shape and a fixed dipole momentum \mathbf{p} . By a fixed dipole we mean that both the absolute value of \mathbf{p} and its orientation with respect to the atom are fixed. The orientation of the vector \mathbf{p} in space can change if the unit rotates with the DA as a whole. Relative positions of the units with respect to each other are assumed fixed due to the bonds of different nature. In Fig. 1b, the bonds are shown schematically as certain specific locks. In our model, the energy accumulated in the locks is formally neglected. The assumption does not mean that this "locking" energy is small (it might, in fact, be huge) but we assume that the "locking" energy per lock is fixed. However, possible change in the total locking energy at the external interface is taken into account. The locking energy



Fig. 1. Macroscopic view (a), microstructure (b), and an elementary cell (c) of a dipole aggregate; ■ – bonds and locks, **p** – dipole vector.

associated with the external interface S can alter due to rearrangement of the nearboundary elementary units. In fact, we assume that the locks can be unlocked by certain energetically favourable fluctuations. Then the "atom" can move along the interface or in its vicinity (or even tunnel through the bulk) and stick to the DA in a new position. In this new position the total accumulated energy is less than that of the original configuration. In other words, the total accumulated energy and the shape of the DA can change substantially due to such a rearrangement. Since both, the total electric energy and the total surface energy, are shape-sensitive, the equilibrium shape corresponds to the balance of the two ingredients.

3. THE EQUILIBRIUM SHAPE OF DIPOLE AGGREGATES

We proceed with a formal presentation of the physical model described above. Conservation of the total mass of the incompressible DA implies the conservation of its total volume V^o in the process of the rearrangement

$$\int_{\Omega} d\Omega = V^o.$$
(2)

The shape of the domain Ω can change due to the rearrangement.

Because of the assumptions made, the dipole polarization vector \mathbf{p} , as well as the polarization \mathbf{P} per unit volume, remain constant in all points of the DA. Since an equilibrium configuration of the isolated DA is defined up to an arbitrary rotation of a DA as a rigid whole, we can assume that the vectors \mathbf{p} and \mathbf{P} are fixed in space and, in particular, they both point in the same direction of a fixed unit spatial vector 1. The total electric energy of the isolated DA in vacuum is equal to

$$E_{el} = -\frac{1}{2} \int_{\Omega} d\Omega \mathbf{P} \mathbf{E} = -\frac{1}{2} \mathbf{P} \int_{\Omega} d\Omega \mathbf{E}, \qquad (3)$$

where **E** is the electric field in the space generated by the dipoles of the DA. In one case, the z^i components of the field **E** in any Cartesian (or affine) coordinate system can be presented by the following explicit formula:

$$E_i(z^k) = P^j \nabla_i \nabla_j \int_{\Omega} d\Omega^* \frac{1}{|\mathbf{z} - \mathbf{z}^*|} = P^j \nabla_i \int_{S} dS^* N_j^* \frac{1}{|\mathbf{z} - \mathbf{z}^*|}.$$
 (4)

Integrations in (4) are with respect to the coordinates z^{*i} , N_j^* is the outward unit normal to the surface S. In the following we use standard rules and notations of tensor calculus (see, for instance, [¹³]): the Latin (spatial) indexes run the values 1, 2, 3; the Greek indexes run the values 1, 2 and are related to the surface coordinates on S; ∇_i and ∇_{α} are the corresponding symbols of covariant differentiation.

Since the field **E** is shape-sensitive, the total electric energy is a shapesensitive quantity too. The potential $\varphi(z)$ of the electric field satisfies the following equations:

1) inside and outside the DA

$$\nabla^i \nabla_i \varphi = 0, \tag{5}$$

2) across the interface S

$$[\varphi]_{-}^{+} = 0, \tag{6}$$

$$[D_i]^+_{-} N^i = 0, (7)$$

3) at infinity

$$\varphi(\infty) = 0, \tag{8}$$

where $D_i = E_i + 4\pi P_i$ is the electric induction and $[A]^+_-$ is the jump of the field A across the interface S.

The specific energy σ per unit area of the surface S can be chosen to be a positive constant, so that

$$E_s = \int\limits_{S} dS\sigma. \tag{9}$$

Hence, we arrive at the problem of minimization of the functional E_{ac} subject to an isoperimetric constraint of the total volume conservation (2), where both the energy and the constraint depend on the shape of the DA.

We conclude that all the non-trivial variations in our variational problem are the normal displacements C of the boundary S. Somewhat lengthy calculations, which cannot be reproduced here (they are quite similar to those in [¹⁴]), lead to the following expression of the first energy variation of the functional $E^* = E_{ac} - \Lambda \int_{\Omega} d\Omega$:

$$\delta E^* = -\int\limits_{S} dSC \left(\sigma B^{\alpha}_{\alpha} + P^k E_{k+} + 2\pi P_i P_j N^i N^j + \Lambda\right), \tag{10}$$

where $B^{\alpha\beta}$ is the tensor of the second quadratic form of the interface S (see, for instance, [¹³]), Λ is the Lagrange multiplier for the volume conservation constraint (2), and E_{k+} is the limit of the electric field as we approach the surface S of the DA from inside.

The choice of the displacements C in different points of the interface S is arbitrary. We arrive at the following equation for possible equilibrium shapes of the DA

$$\sigma B^{\alpha}_{\alpha} + P^k E_{k+} + 2\pi P_i P_j N^i N^j = -\Lambda. \tag{11}$$

In addition to Eq. (11), the full system of equilibrium equations includes also Eqs. (2) and (5)–(8). It is clear from physics that at least one of the solutions of the system should resemble a spheroid elongated in the l-direction (the dipole energy is the least if all dipoles form a line whereas surface energy will be least for the spherical shape). That is probably why Heisenberg assumes that ferromagnetic domains take on the needle-like shape. Landau and Lifshits [³] do not share this opinion, though. So far, the author has not been able to find analytically the equilibrium shape of a finite DA. However, there are certainly no exact solutions: 1) an infinite lamella with plane parallel boundaries and arbitrarily oriented polarization vector, and 2) an infinite circular cylindrical tube with the coaxial polarization vector \mathbf{P} . Despite their "unboundedness", these solutions are certainly instructive for understanding various features of DA. We discuss below how to calculate possible equilibrium shapes of the DA numerically.

4. STABILITY OF DIPOLE AGGREGATES

In order to study the stability of an equilibrium shape of DA, we calculate the second energy variation. Relevant calculations (similar to those of Chapter 4 of $[^{14}]$) lead to the following formula:

$$\delta^{2}E^{*} = -\sigma \int_{S} dSC \left(\nabla^{\alpha}\nabla_{\alpha}C + CB^{\alpha}_{\beta}B^{\beta}_{\alpha} \right) \\ + \int_{S} dSC \left(-P^{k}\delta E_{k+} + 4\pi P^{i}P^{j}N_{i}z_{j}^{\cdot\alpha}\nabla_{\alpha}C - CN^{l}P^{k}\nabla_{l}E_{k+} \right), (12)$$

where $z_{j.}^{\alpha}$ is the shift tensor of the boundary (see [^{13,14}]) and δE_i is the variation of the field E_i caused by variation of equilibrium position of the boundary S.

The problem of morphological stability of the interface with respect to the short wavelength variations, localized close the interface variations, is much easier than the general stability problem and much more interesting from the standpoint of physics. In fact, if the near-boundary disturbances are much shorter than the radii of curvature of the boundary and the spatial inhomogeneity of the DA, one can safely limit himself to the study of the stability of the uniform half-space. In this situation the system is translationally invariant in the in-plane directions, and the Fourier analysis of the second energy variation gives all required information on the necessary conditions of morphological stability.

Realization of this programme results in the following formula of the second energy variation in terms of the Fourier-components $C^*(\mathbf{k})$ of the interface disturbances with the in-plane wave vector \mathbf{k} :

$$\delta^2 E^* = \int_{R^2} d^2 \mathbf{k} C^*(-\mathbf{k}) C^*(\mathbf{k}) \left| \mathbf{k} \right| \left\{ \sigma |\mathbf{k}| + 2\pi \left(\frac{1}{\left| \mathbf{k} \right|^2} \left(\mathbf{k} \cdot \mathbf{P} \right)^2 - (\mathbf{N} \cdot \mathbf{P})^2 \right) \right\}.$$
(13)

Equation (13) shows that the normal component \mathbf{P}_{\perp} of the polarization vector \mathbf{P} destabilizes the interface, whereas the tangential component \mathbf{P}_{\parallel} stabilizes it. The interface is absolutely stable provided the tangential component is greater than the normal one:

$$\left|\mathbf{P}_{\parallel}\right| \ge \left|\mathbf{P}_{\perp}\right|. \tag{14}$$

If the opposite inequality is true, then morphological stability is guaranteed for sufficiently short surface rearrangements. The long surface perturbations are still unstable. The critical (neutral) in-plane wave vectors \mathbf{k}_{crit} satisfy the following formula:

$$\frac{\sigma}{2\pi} |\mathbf{k}_{crit}| = (\mathbf{e} \cdot \mathbf{P})^2 - (\mathbf{N} \cdot \mathbf{P})^2, \qquad (15)$$

where e is unit in-plane vector which is collinear with the vector k.

We turn now to the stability of a DA in the shape of an infinite cylinder with axially directed dipoles. The axial electric field partially stabilizes the well-known Plateau–Rayleigh morphological instability with respect to the sufficiently lengthy axisymmetric shape variations. We denote by R the equilibrium radius of the cylinder and by k the axial wavenumber of the disturbances. Using the formula of the second energy variation (12), we arrive at the following equation binding the neutral (critical) disturbance parameter $\zeta = k_{ne}R$ and the dimensionless ratio $\Gamma = 4\pi P^2 R/\sigma$ (describing the trade-off between the DA-dipole and the DA-surface energies):

$$\left(1 - \frac{1}{\varsigma^2}\right)\varsigma \frac{d}{d\varsigma} \left(\ln \frac{I_0(\varsigma)}{K_0(\varsigma)}\right) + \Gamma = 0, \tag{16}$$

where I_0 and K_0 are the zero-order Bessel functions of imaginary argument bounded at $\varsigma = 0$ and $\varsigma = \infty$, respectively. Any standard software allows one to plot the root $\varsigma(\Gamma)$ of Eq. (16) and to see the growth of the critical wavelength when Γ increases.

In the context of astro- and geophysical hydrodynamics, the axial magnetic field stabilization of the self-gravitating liquid cylinders was first investigated by Chandrasekhar and Fermi [¹⁵].

5. SLOW EVOLUTION OF DIPOLE AGGREGATES

In this section we present the system allowing one to calculate possible equilibrium shapes of DA using the technique of surface-diffusion-minimization in the spirit of the onsageristic linear irreversible thermodynamics.

In order to satisfy the mass conservation constraint (2), we choose C in the form of the surface divergence $\nabla_{\alpha} J^{\alpha}$ of the surface vector field J^{α} (which some people are inclined to interpret as the surface flux of the substance)

$$C = \nabla_{\beta} J^{\beta}. \tag{17}$$

The decay of the total energy along the trajectory is obviously guaranteed if one chooses the following constitutive equation of J^{β}

$$J^{\beta} = -\varkappa \nabla^{\beta} \left(\sigma B^{\alpha}_{\alpha} + P^{k} E_{k+} + 2\pi P_{i} P_{j} N^{i} N^{j} \right), \tag{18}$$

where \varkappa is a positive constant. Excluding J^{β} between (17) and (18), we get

$$C = -\varkappa \nabla_{\beta} \nabla^{\beta} \left(\sigma B^{\alpha}_{\alpha} + P^{k} E_{k+} + 2\pi P_{i} P_{j} N^{i} N^{j} \right).$$
⁽¹⁹⁾

This technique allows one to find equilibrium shapes of DAs starting with any configuration satisfying the constraint (2). For this configuration one has to solve the standard electrostatic system (5)–(8) and then to calculate the surface scalar field C using Eq. (19). The initial shape should be, then, upgraded by "moving" the boundary along the outward normal N^i for the distance C. Iterating the procedure with the upgraded shape one can find the (stable!) equilibrium shapes of DA satisfying the "chemical" condition (11).

Not all researchers are inclined to accept the linear irreversible thermodynamics and the theory of surface diffusion, in particular. Needless to emphasize though, that regardless of its validity from the standpoint of physics, this technique allows one to find possible equilibrium shapes of DAs numerically. The calculated equilibrium shapes are certainly sensitive to the choice of the initial configuration. There is no need to struggle against this non-uniqueness, it just reflects the nature of the problem in question.

6. CONCLUSIONS

We have analysed some aspects of thermodynamic basics of equilibrium shapes and stability of aggregates with dipole electrostatic or magnetostatic interactions. In order to calculate the equilibrium shape of the simplest model of DA, described in Section 2, one has to solve the standard electrostatic system of equations (5)–(8)subject to the mass conservation constraint (2) and the condition of equilibrium with respect to mass rearrangement (11). The morphological stability of DA can be analysed by means of the second energy variation given by Eq. (12). In particular, we established that, even in the absence of the surface energy stabilization the interface of DA is morphologically stable with respect to any local rearrangement, provided that the tangential component of the dipole momentum vector is greater than the normal component of that vector. Otherwise the wave vector of the stable corrugations should be greater than \mathbf{k}_{crit} defined by Eq. (15) and depending on the surface energy density and on the orientation of surface corrugations. The local morphologic stability of the interface in each point does not guarantee the stability of the DA in whole. For instance, the tangential orientation of the dipole electric field guarantees the local stability of the interface. However, a DA in the shape of an infinite cylinder is stable with respect to sufficiently lengthy corrugation: Eq. (16) allows calculation of the critical wavelength of the radially symmetric corrugations.

The equilibrium shape of the DA can be calculated numerically with the help of an iteration procedure based on the system (5)–(8) augmented with the evolution equation (19). I hope that this system can be used as a convenient tool not only in the experimental study of the equilibrium shapes of DAs but also in the study of morphogenesis of DA caused by slow evolution of, say, surface energy or of certain external parameters of the media. Some modifications of the master system are required for such considerations. Among numerous problems which can be addressed by means of the evolutionary system formulated above, the following are the most intricating from physical and mathematical viewpoints.

1. What are possible equilibrium shapes of an isolated DA?

2. How many different equilibrium shapes of an isolated DA do exist? Are they necessarily axisymmetric?

3. Is an isolated equilibrium DA necessarily homomorphic to a sphere? In particular, is there an equilibrium toroidal isolated DA?

4. Is there a maximal (critical) volume of an isolated DA?

5. Can the equilibrium shape of an isolated DA bifurcate?

The distance between the theory of electro/magnetostatics of DA and of a dynamic theory of DA is obviously much greater than the distance between the statics of Archimedes and the dynamics of Newton. But if the "diffusion-like" minimization, described in the last section of this paper, is capable of describing certain *real evolutionary processes* of DAs, the above list can be extended with the following problems.

6. Can two DAs collide and how do their shapes evolve in the process of colliding?

7. Does the Ostwald ripening take place for DAs?

Regardless of its ability to describe any real physical *process*, the consideration of the "diffusion" system presented above is a useful step in the development of the computation-visualization methods in the physics of the DA morphogenesis. In particular, it is instrumental in answering all the above questions by means of computational experimentation.

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DIPOOLAGREGAATIDE MORFOLOOGIA, STABIILSUS JA MORFOGENEES

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On käsitletud lihtsate dipolaarsete süsteemide teooriat. Uuritav objekt – dipoolagregaat – on võimeline akumuleerima elektri- või magnetostaatilist energiat ning samuti pinnaenergiat, kuid mitte siseenergiat. On esitatud dipoolagregaatide analüüsiks ja numbriliseks simuleerimiseks vajalikud võrrandisüsteemid ja termodünaamika seosed, mis võimaldavad määrata nende tasakaaluseisundeid ja kvaasistaatilist evolutsiooni.