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Relaxation of interstitials in spherical colloidal crystals

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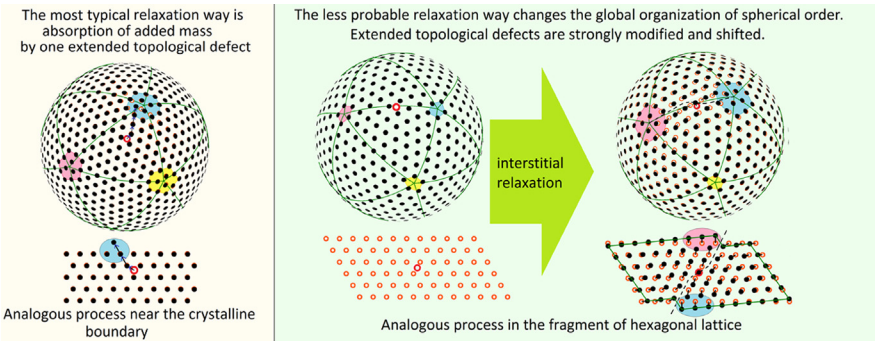
HIGHLIGHTS

- Ways of interstitials relaxation in spherical colloidal crystals are classified.
- The most probable relaxation way involves only one extended topological defect.
- Parent phase reveals changes in global organization of relaxed hexagonal order.
- Interstitials fractionalization is analyzed in terms of parent icosahedron method.
- Relationships between relaxation in planar and spherical crystals are found.

GRAPHICAL ABSTRACT



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ABSTRACT

Spherical colloidal crystals (CCs) self-assemble on the interface between two liquids. These 2D structures unconventionally combine local hexagonal order and spherical geometry. Nowadays CCs are actively studied by altering their structures. However, the statistical analysis of such experiments results is limited by uniqueness of self-assembled structures and their short lifetime. Here we perform numerical experiments to investigate pathways of CC structure relaxation after the intrusion of interstitial. The process is simulated in the frames of overdamped molecular dynamics method. The relaxation occurs due to interaction with extended topological defects (ETDs) mandatory induced in spherical CCs by their intrinsic Gaussian curvature. Types of relaxation pathways are classified and their probabilities are estimated in the low-temperature region. To analyze the structural changes during the relaxation we use a parent phase approach allowing us to describe the global organization of spherical order. This organization is preserved by only the most typical relaxation pathway resulting in filling one of vacancies integrated inside the ETD areas. In contrast with this pathway the other ones shift the ETDs centers and can strongly reconstruct the internal structure of ETDs. Temperature dependence of the relaxation processes and the mechanism of dislocation unbinding are discussed. Common peculiarities in relaxation of spherical structures and particular fragments of planar hexagonal lattice are found.

1. Introduction

The fact that colloidal particles can be adsorbed at the interface between two liquids has been known since the beginning of the 20th century [1]. However, the first colloidal crystals on non-planar surfaces were obtained only recently [2]. These 2D objects are

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characterized by a local hexagonal order and the obligatory presence of topological defects induced by non-zero Gaussian curvature [3–5]. Topological defects occupy the extended areas on the surface of colloidal crystals (CCs). In these regions a local hexagonal order is significantly distorted. If the defect area has non-zero topological charge [4] the defect is called a scar [4,6], otherwise a pleat [7,8]. The presence of 12 scar-like extended topological defects (ETDs) is typical of the spherical CCs [3–6]. The centers of these ETDs are approximately equidistant from each other, so they are located near the vertices of the icosahedron [3]. Pleats are usually observed in the CCs which are self-assembled on the surfaces with negative Gaussian curvature [7–9].

Over the last decade experimental methods for studying 2D CCs were substantially advanced. Previously, the CC structures were simply observed with an optical microscope [2,4]. Now it is possible to study these objects by active altering their structure with the new experimental methods, which originate from the optical tweezers technique. Use of these methods allows moving individual colloidal particles [8,10] or coherent changing the positions of the whole groups of them [8,10]. After the enforced reconstructions colloidal structure relaxes. This process can be directly observed and registered using an optical microscope.

It is especially interesting to observe the relaxation of the interstitials intruded into spherical CCs [8], where the explicit crystal boundary is absent. After the intrusion of an extra particle, the local hexagonal order is restored around it and one or few nearest ETDs are reconstructed. As is well known, the interstitial close to the crystal boundary can relax due to a shift of lattice nodes towards the crystal boundary [11–13]. In this case the last particle of the shifted line is pushed upon the crystal surface. Such a process can be considered as a movement of two dislocations [11–13] with zero total Burgers vector to the crystal boundary. Relaxation of the interstitial in the spherical CCs occurs similarly since the latter process also represents the movement of a few dislocations [8,14], which are absorbed by the closest ETDs. The well-known interpretation of scars as grain boundaries [4] allows us to understand that the dislocation absorption by scar is analogous to the process of its absorption by the crystal surface [8]. In both cases, the dislocation charge is not conserved [15].

The crystal boundary can absorb an unlimited number of dislocations. In contrast, the scar is always finite, and can be always characterized by a small *discrete* number of vacancies embedded into its structure [15]. Nevertheless, during the relaxation the interstitial can be '*fractionalized*' [8]. In other words the additional mass of the single intruded particle is redistributed over the spherical crystal. Here we study in details this process and show that such mass splitting is only one side of the interstitial relaxation process. The other side is the change in the global organization of spherical hexagonal order. This organization is analyzed in terms of a parent icosahedron method that is proposed in this article.

There are a large number of works devoted to the relaxation of interstitials and vacancies in flat [8,16] and spherical [8,10,14] crystals. However, as far as we know, the defect relaxation has never been considered in terms of preserving or changing the global organization of the hexagonal order on the sphere. Here we develop the parent phase approach and demonstrate how the global order reconstruction occurs while several ETDs simultaneously participate in the relaxation of interstitial. The article also clarifies analogies between the relaxation processes occurring in spherical CCs and those, which take place in the finite fragments of an ordinary 2D hexagonal lattice.

2. Theoretical methods

In this section we recall some known properties of the low-energy spherical hexagonal order. To characterize the processes of the interstitial relaxation we introduce a concept of global organization of such spherical order. After that we consider the overdamped dynamics of colloidal particles and apply it to model the interstitial relaxation.

2.1. Low-energy hexagonal order on the sphere and the parent icosahedron approach

The particles in the colloidal crystal are retained by surface tension at the interface between the two liquids and interact each with other with screened Coulomb potential [8]. Ordinarily the interaction potential is simplified and replaced by conventional Coulomb-like expression corresponding to the particles repulsion. Thus, the order in the spherical CCs is successfully modeled and analytically studied [17–19] in the frame of the simplest electrostatic energy:

$$U = \sum_{j>i}^{N} \frac{1}{r_{ij}}, \tag{1}$$

where r_{ij} is the distance between ith and jth particles and N is the total number of particles. The self-assembly of 2D structure on a curved surface is described by the conditional minimization of the energy (1) with respect to coordinates of the particles. The condition imposed is that any particle during minimization should be retained on the surface under consideration.

Structures that for a given N correspond to the global minimum (1) are the solutions of the Thomson problems [20–24]. The equilibrium energies (1) of structures corresponding to global and local-minima are very close. Moreover, the difference between the equilibrium energies (1) is strongly reduced and the number of equilibrium structures grows exponentially with the number N of particles in the structure [25]. Therefore, the structure of the real CC would rather correspond to one of the numerous deep local minima of energy (1), than to exact solution of the Thomson problem. However, since the solutions of Thomson problems are well known from the literature it is more convenient to use them to model the order in spherical CCs. Therefore here we study the interstitial relaxations on examples of model spherical structures (MSSs) corresponding to the deepest known today minima of energy (1).

Now let us recall some unusual properties of the hexagonal order on the sphere [5,13]. Sufficiently large lowest energy MSSs with $N \geq 400$ have 12 scar-like ETDs [3,23] which are located near the vertices of regular or slightly distorted spherical icosahedron covered by a simply connected hexagonal lattice. Formation of ETDs in the spherical crystals is induced by the Gaussian curvature of the spherical surface. The hexagonal order in the defect areas is strongly distorted but any defect can be surrounded by a pentagonal contour with sides being parallel to the minimal translations of the order [15]. The pentagonal shape of the surrounding contour [5,15] is caused by the fact that each ETD has the minimal positive total topological charge $+1$.

Below we apply a parent icosahedron (PI) method to characterize the global organization of the hexagonal order on the sphere. So let us describe the PI construction briefly. Triangulation of the order inside the ETD reveals the linear scar [4], which is a chain of alternating particles with 5 or 7 nearest neighbors. Virtual insert of a small number N_{ins} of particles inside the pentagonal contour surrounding the ETD allows the topological reconstruction of the order inside the defect [15]. In the result of this procedure all particles inside the defect area except the single one obtain six neighbors and the scar disappears [15]. Location of the appearing 5-fold disclination is completely determined by the location and shape of the pentagonal contour surrounding the initial ETD [15].