様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科学研究費助成事業　　研究成果報告書

令和　６ 年　 ４ 月 ２６ 日現在

機関番号：１１３０１
研究種目：若手研究
研究期間：2020～2023
課題番号：２０Ｋ１４４３７

研究課題名（和文）Effect of patchy particle designs on the bulk properties of the self-assembled structures

研究課題名（英文）Effect of patchy particle designs on the bulk properties of the self-assembled structures

研究代表者
Ｌｉｅｕ　Ｕｙｅｎ（Lieu, Uyen）

東北大学・材料科学高等研究所・助教（研究特任）

研究者番号：００８０７０４２

交付決定額（研究期間全体）：（直接経費）　　1,800,000円

研究成果の概要（和文）：私たちは、パッチ粒子の自己組織化についていくつかの側面を調査してきました。（i）私たちは、球面上の双極子状パッチ粒子の自己組織化において、位相欠陥がどのように現れるかを明らかにしました。（ii）パッチ粒子から組み立てられた十二角形の準結晶の成長と揺らぎのメカニズムが明らかにされています。（iii）与えられた構造に対して、最適なパッチ粒子の種類を見つける逆設計アルゴリズムを提案しています。（iv）パッチ粒子と所望の構造が与えられた場合、自己組織化中に所望の構造が最も速く効率的に得られるように、最適な制御パラメータを学習するために強化学習を使用することを提案しています。

研究成果の学術的意義や社会的意義
The research shows that the self-assembly of nano- and colloidal particles is promising in the development of new materials. We have proposed simulation methods that help to find the best way to obtain desired structures.

研究成果の概要（英文）：We have investigated the self-assembly of patchy particles on several aspects. (i) We have elucidate how the topological defects appear on the self-assembly of dipole-like patchy particle on a spherical surface. (ii) The mechanism of the growth and fluctuation of a dodecagonal quasicrystal assembled from patchy particles are revealed. (iii) We propose an inverse design algorithm so that for a given structure, the best type of patchy particle can be found out. (iv) Given the patchy particle and the desired structure, we propose to use reinforcement learning to learn the best control parameter during the self-assembly so that the desired structure is obtained fastest and most efficient.

研究分野：生物物理、化学物理およびソフトマターの物理関連

キーワード：Patchy particle　Self-assembly　Mechanism　Structure　Reinforcement learning　Inverse design

様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９（共通）

１．研究開始当初の背景

The search for novel materials, which is fundamentally different from the conventional one, is the key element for the development of new materials. One of the highlighting candidates is colloidal particles from which a micro-crystal is built up. The properties of the crystal are determined by the properties of the particles in the structure and their interaction. The transfer of light, heat, etc. in these crystals can be manipulated to obtain special optical, thermal properties. Patchy particles, the particles with anisotropic surface patterns, are of great interest in colloidal sciences. Patchy particles have patches on specific places on the surface, making the interaction of such particles not only dependent on the distance, but also on their mutual orientation. Therefore, the patchy particles can self-organise into much more complex structure than the case of isotropic particles. The degree of freedoms (i.e. anisotropy) of the particle have direct impact on the possible outcome structure. Although there are infinitive number of designs for particle anisotropy (Figure 1), it is difficult to predict the suitable patchy particle for a specific structure and vice versa, even for the case of spherical particle (with non-spherical pattern). We investigate this issue by numerical simulation.

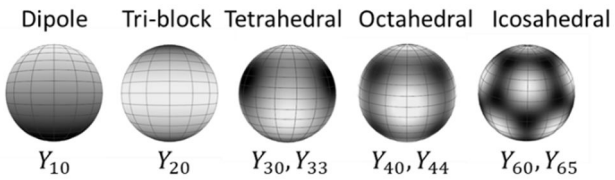

Figure 1: Examples of patterns on patchy particles in by using spherical harmonics Y_{lm} and their combinations

２．研究の目的

The purpose of this research is to investigate how the anisotropy of the patchy particle affects the structures and vice versa. We consider the following aspects:

(1) For a given type of patchy particle, what is the possible assembled structure in different condition? The effect of curvature on self-assembly is investigated. Moreover, we study the growth of a quasicrystalline structure by patchy particles.

(2) For a desired assembled structure, how to know the possible design of the particle?

(3) When the design of the particle is known, how to optimize the operational parameter during the assembly to maximise the yield, can we utilize a data-driven model so that the model knows how to adjust the parameter on-the-fly?

３．研究の方法

Here is the core of the method of research:

(1) Model for anisotropy of the patchy particle is based on spherical harmonics. The interaction of a pair of particles is estimated by Taylor expansion and irreducible tensor.

(2) The dynamics of the particles is described by Brownian dynamics simulation.

(3) The self-assembled structure is analysed by several parameters, such as bond-orientational order parameters, fast Fourier transformation.

(4) The inverse design is proposed from the relative entropy method. From the framework for isotropic particle system, we have derived the model for the anisotropic particle, in which the orientation of the particle has to be taken into account.

(5) As for the reinforcement learning, we use the simple Q-learning algorithm.

４．研究成果

(1) We investigate the assembly of dipole-like patchy particles confined to a spherical surface by Brownian dynamics simulations. The surface property of the spherical particle is described by the spherical harmonic Y_{10}, and the orientation of the particle is defined as the uniaxial axis. On a flat space, we observe a defect-free square lattice with nematic order. On a spherical surface, defects appear due to the topological constraint (Figure 2). As for the director field, four defects of winding number +1/2 are observed, satisfying the Euler characteristic. We have found many configurations of the four defects lying near a great circle. Regarding the positional order for the square lattice, eight grain boundary scars proliferate linearly with the sphere size. The positions and orientations of the eight grain boundary scars are strongly related to the four +1/2 defect cores.

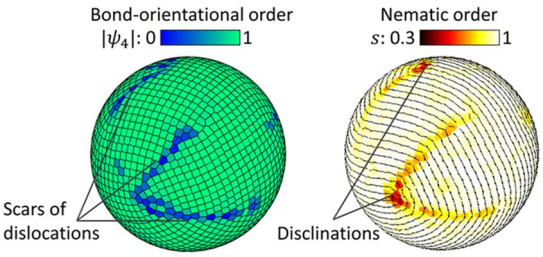

Figure 2. Dipole particles on a sphere. Particles occupy Voronoi cells (left) and directors (right).

(2) The self-assembly of two-dimensional dodecagonal quasicrystals (DDQCs) from patchy particles is investigated by Brownian dynamics simulations. The patchy particle has a five-fold rotational symmetry pattern described by the spherical harmonics Y_{55}. From the formation of the DDQC obtained by an annealing process, we find the following mechanism. The early stage of dynamics is dominated by hexagonal structures. Then, nucleation of dodecagonal motifs appears by particle rearrangement, and finally the motifs span the whole system. The transition from the hexagonal structure into the dodecagonal motif coincides with the collective motion of the particles. The DDQC consists of clusters of dodecagonal motifs, which can be classified into several packing structures (Figure 3). By the analyses of the DDQC under fixed temperature, we find that the fluctuations are characterised by changes in the network of the dodecagonal motifs. Finally, we compare the DDQCs assembled from the patchy particle system and isotropic particle system. The two systems share a similar mechanism of the formation and fluctuation of DDQCs.

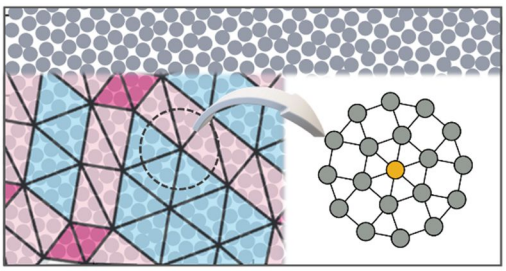

Figure 3. The order of the particles in a dodecagonal quasicrystal after decorating the particles according to their local structures and the triangulated network.

(3) We propose an optimisation method for the inverse structural design of self-assembly of anisotropic patchy particles. The anisotropic interaction can be expressed by the spherical harmonics of the surface pattern on a patchy particle, and thus arbitrary symmetry of the patch can be treated. The pairwise interaction potential includes several to-be-optimised parameters, which are the coefficient of each term in the spherical harmonics. We use the optimisation method based on the relative entropy approach and generate structures by Brownian Dynamics simulations. Our method successfully estimates the parameters in the potential for the target structures, such as square lattice, kagome lattice, and dodecagonal quasicrystal.

(4) We propose reinforcement learning to control the dynamical self-assembly of the dodecagonal quasicrystal (DDQC) from patchy particles. The patchy particles have anisotropic interactions with other particles and form DDQC. However, their structures at steady states are significantly influenced by the kinetic pathways of their structural formation. We estimate the best policy of temperature control trained by the Q-learning method and demonstrate that we can generate DDQC with few defects using the estimated policy. The temperature schedule obtained by reinforcement learning can reproduce the desired structure more efficiently than the conventional pre-fixed temperature schedule, such as annealing. To clarify the success of the learning, we also analyse a simple model describing the kinetics of structural changes through the motion in a triple-well potential. We have found that reinforcement learning autonomously discovers the critical temperature at which structural fluctuations enhance the chance of forming a globally stable state. The estimated policy guides the system toward the critical temperature to assist the formation of DDQC.

５．主な発表論文等

〔雑誌論文〕　計3件（うち査読付論文　3件／うち国際共著　3件／うちオープンアクセス　3件）

１．著者名 Lieu Uyen Tu、Yoshinaga Natsuhiko	４．巻 18
２．論文標題 Formation and fluctuation of two-dimensional dodecagonal quasicrystals	５．発行年 2022年
３．雑誌名 Soft Matter	６．最初と最後の頁 7497～7509
掲載論文のDOI（デジタルオブジェクト識別子） 10.1039/D2SM00798C	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Lieu Uyen Tu、Yoshinaga Natsuhiko	４．巻 156
２．論文標題 Inverse design of two-dimensional structure by self-assembly of patchy particles	５．発行年 2022年
３．雑誌名 The Journal of Chemical Physics	６．最初と最後の頁 054901～054901
掲載論文のDOI（デジタルオブジェクト識別子） 10.1063/5.0072234	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Lieu Uyen Tu、Yoshinaga Natsuhiko	４．巻 16
２．論文標題 Topological defects of dipole patchy particles on a spherical surface	５．発行年 2020年
３．雑誌名 Soft Matter	６．最初と最後の頁 7667～7675
掲載論文のDOI（デジタルオブジェクト識別子） 10.1039/D0SM00103A	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

〔学会発表〕　計12件（うち招待講演　0件／うち国際学会　11件）

１．発表者名 Uyen Tu Lieu, Natsuhiko Yoshinaga
２．発表標題 Reinforcement learning for dynamic control of self-assembly of quasicrystals from patchy particles
３．学会等名 APS March Meeting 2024（国際学会）
４．発表年 2024年

１．発表者名
Uyen Lieu, Natsuhiko Yoshinaga

２．発表標題
Reinforcement learning for dynamic control of self-assembly of quasicrystals from patchy particles

３．学会等名
28th Quasicrystals Meeting (Hypermaterials)

４．発表年
2023年

１．発表者名
Uyen Tu Lieu, Natsuhiko Yoshinaga

２．発表標題
Inverse Design of Two-dimensional Dodecagonal Quasicrystal Structure by Patchy Particles

３．学会等名
7th International Soft Matter Conference (ISMC2023)（国際学会）

４．発表年
2023年

１．発表者名
Uyen Lieu, Natsuhiko Yoshinaga

２．発表標題
Inverse design of two-dimensional dodecagonal quasicrystal structure by patchy particles

３．学会等名
28th International Conference on Statistical Physics (Statphys28)（国際学会）

４．発表年
2023年

１．発表者名
Uyen Lieu, Natsuhiko Yoshinaga

２．発表標題
Inverse Design of Two-dimensional Dodecagonal Quasicrystal Structure by Patchy Particles

３．学会等名
KITP Conference "Structure Design and Emerging Phenomena in Nanoparticle Assemblies: What ' s Next? "（国際学会）

４．発表年
2023年

１．発表者名
Uyen Lieu

２．発表標題
Simulation of self-assembly of a two-dimensional dodecagonal quasicrystal using patchy particles

３．学会等名
Aperiodic 2022（国際学会）

４．発表年
2022年

１．発表者名
Uyen Lieu

２．発表標題
Inverse Design of Two-dimensional Self-assembly of Patchy Particles

３．学会等名
APS March Meeting 2022（国際学会）

４．発表年
2022年

１．発表者名
Uyen Lieu

２．発表標題
Inverse Design of Two-dimensional Self-assembly of Patchy Particles

３．学会等名
Material Research Meeting 2021 (MRM2021)（国際学会）

４．発表年
2021年

１．発表者名
Uyen Lieu

２．発表標題
Inverse Design of Two-dimensional Self-assembly of Patchy Particles

３．学会等名
IRN-Aperiodic 2021（国際学会）

４．発表年
2021年

１．発表者名
Uyen Lieu

２．発表標題
Inverse Design of Two-dimensional Self-assembly of Patchy Particles

３．学会等名
XXXII IUPAP Conference on Computational Physics（国際学会）

４．発表年
2021年

１．発表者名
Uyen Lieu

２．発表標題
Topological defects of patchy particles on a spherical surface

３．学会等名
11th Liquid Matter Conference (LMC2020/2021)（国際学会）

４．発表年
2021年

１．発表者名
Uyen Lieu

２．発表標題
Inverse Design of Patchy Particle for Self-assembly

３．学会等名
1st International School on Hypermaterials (ISH2021)（国際学会）

４．発表年
2021年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

―

６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考
研究協力者	義永　那津人 (Yoshinaga Natsuhiko)		

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関