RGC Ref.: A-HKUST616/14
(please insert ref. above)

The Research Grants Council of Hong Kong
ANR/RGC Joint Research Scheme
Completion Report

(Please attach a copy of the completion report submitted to the ANR by the French researcher)

Part A: The Project and Investigator(s)

1. Project Title (ANR Acronym)

Assembly and dynamics of active and passive micro-ellipsoids at a fluid interface

2. Investigator(s) and Academic Department/Units Involved

	Hong Kong Team	French Team
Name of Principal Investigator *(with title)*	Prof Yilong Han	Prof Maurizio Nobili
Post	professor	Professor
Unit / Department / Institution	Department of Physics /Hong Kong University of Science and Technology	Laboratoire Charles Coulomb, UMR5521 Université Montpellier 2/CNRS
Contact Information	yilong@ust.hk	maurizio.nobili@umontpellier.fr
Co-investigator(s) *(with title and institution)*	Penger Tong, Chair Professor Department of Physics /Hong Kong University of Science and Technology	Christophe Blanc, Experienced Scientist Antonio Stocco, Junior Scientist Martin In, Senior Scientist Michel Gross, Senior Scientist Laboratoire Charles Coulomb, UMR5521 Université Montpellier 2/CNRS

3. Project Duration

	Original	Revised	Date of RGC/ Institution Approval *(must be quoted)*
Project Start date	Apr.1, 2015	Mar.1, 2015	Jan.23, 2015
Project Completion date	Mar.31, 2019	Feb.28, 2019	
Duration *(in month)*	48	48	

Deadline for Submission of Completion Report	Mar. 31, 2020	Feb.28, 2020	

<u>Part B: The Completion Report</u>

5. Project Objectives

5.1 Objectives as per original application

1. The first objective is the fabrication of ellipsoids and elliptical platelets with tunable sizes, aspect ratios, activity, physico-chemical properties and surface heterogeneities. The fabrication of such particles will be coupled with the interface design by an appropriate choice of the fluids concerned and of the interface curvature in order to control the capillary attraction between particles down to the thermal level. This will allow a major scientific breakthrough as it will open the way to the study of 2D concentrated systems of anisotropic particle without the actual limitation due to the strong capillary attraction.

2. The second objective which concerns anisotropic passive particles, is to investigate the interface breaching dynamics and the 2D particle diffusion dynamics at the interface. To this end at L2C we will develop a new digital holography based on FDTD (Finite-difference time-domain method) applicable to interfaces separating fluids with different optical indices and giving directly the particle position and orientation with respect to the interface. We will further develop the hanging fiber AFM to measure the force during the particle interface breaching.

3. The third objective is to characterize the particle interactions with the fluid interface, its dynamics close to the interface and how these phenomena depend on the particles properties and on the physico-chemistry of the interface. To afford this study an integrated optical system coupling multi-trap optical tweezers and digital holography will be build-up at L2C.

4. The fourth objective is to address the motion of anisotropic active colloids both close to the interface and over the interface in order to quantify the fluctuations of the driving force on the particle due to the interface presence. Such measurements will be the first in the field of active colloids.

5. The fifth objective is to characterize collective behavior, the rotator phase and glassy transitions in concentrated 2D systems of anisotropic particles. Some basic problems will be studied, including the frustration in rotator phase and the novel structural signatures in glass transitions.

5.2 Revised Objectives

Date of approval from the RGC: ____________________

Reasons for the change: ____

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1.
2.
3. ....

6. Research Outcome

Major findings and research outcome
(maximum 1 page; please make reference to Part C where necessary)

We have fabricated various types of ellipsoids and develop some cutting-edge measurement techniques including the digital holography microscopy, interferometry near liquid interfaces and liquid-phase AFM for interfacial force measurement. We measured the tiny force between ellipsoids and air-water interfaces under various conditions and found two novel equilibrium positions with peculiar different dynamics along the tangential and normal directions. We discovered four types of dynamics during the particle approaches to the interface. Some of the experimental observations are explained by our theoretical models, while others need future studies. Besides these results mainly contributed by the French group, we played a major role in the following studies.

"What is the nature of glass transition?" has been ranked as one of the 125 major questions in science by journal "Science". The answer remains elusive after intensive studied. We believe that our results provide a major advance to answer this question, and the manuscript will soon be submitted to Nature Materials. We performed comprehensive experiments under different parameters and found the universal Ising critical behavior for both structure and dynamics at the ideal glass transition point and another critical behavior only for dynamics at the model coupling critical point. Hence it is a thermodynamic transition, while there is another pure dynamic transition at the mode-coupling point. Numerous papers on top journals reported various structure-dynamic relations to demonstrate the thermodynamic nature of the transition, but these features cannot be quantitatively checked and thus are much less convincing than the critical behaviors. Ising critical behavior has only been observed in glassy systems with crystalline patches composed of spheres, thus it is not widely accepted although the publication was ranked as the 20 landmark papers in the 10-year history of Nature Materials. Here we also observed the Ising critical behavior with more analysis methods in systems composed of nonspheres (ellipsoids) without any crystalline structure, hence it provides the convincing evidence of the critical behavior. This work is our represented results in the past 10 years

We developed a new technique of long-needle AFM for measuring the tiny capillary force on the solid particle at the liquid-air interface. We found a universal behavior of the asymmetric speed dependence of force hysteresis and contact line relaxation for different microfibers and fluids, and provided a theoretical understanding (Phys. Rev. E 2017, Langmiur 2017)

In addition, we studied the frustration in a buckled colloid monolayer, and discovered rich glassy dynamics. It provides a novel platform to study the glass dynamics in an ordered structure, and explains the puzzle of the observed glassy dynamics in molecular crystals. The results were published in the top physics journal (Phys. Rev. X 2017). Following the direction of the order-disorder transition, we proposed a rarely asked basic question: how to distinguish a fine-grained polycrystal and glass? Surprisingly we found many features jump at one point, indicating a sharp transition rather than a gradual crossover (Phys. Rev. X. 2018). This study open a new approach to study the poorly explored crossover between ultrafine-grain polycrystal and glass. The results can guide the fabrication of novel ultrafine-grained polycrystals which are often unstable and hard to achieve. Another monolayer glass study by us is the first experimental observation at the free surface of a glass with single-article dynamics using our novel colloidal system (Nat. Comm. 2017); We not only found the surface mobile layer, but also discovered another deeper surface layer with distinct properties from the bulk. The dynamics in colloidal glasses arises form the complex energy landscape. We systematical measured the colloidal diffusion on a quasicrystalline substrate and showed how the energy landscape affects the diffusion. We further developed two theoretical models to describe the long-time diffusion of the colloidal particles over a quasicrystal lattice (Soft Matter 2017; J. Chem. Phys. 2017, Editor's pick).

Potential for further development of the research and the proposed course of action *(maximum half a page)*

Since all the team members are experimentalists, one future work for theorists is to provide the theoretical understandings of some of our observations, including the second equilibrium positions for a particle near the interface, the mechanisms of the four types of particle dynamics, the mechanism and nature of the polycrystal-glass transition, and the cause of the Ising critical behavior and another dynamic critical behavior.

In our current preliminary experiment, we added a novel dye-induced tunable attractions between ellipsoids and achieved colloidal glasses composed of tunable attractive nonspherical particles for the first time. This system provide a powerful platform for the study of glasses, including the surface behaviors of attractive glasses and the ultrastable glasses (i.e. its melting and vibration mode studies) with the resolution of single-particle dynamics for the first time. These two active topics in glass studies still lack of experiment with the single-particle dynamics.

We developed advanced measurement techniques including the digital holography microscopy, interferometry near liquid interfaces and liquid-phase AFM for interfacial force measurement. They can be applied to various studies about particles at interfaces and force measurement, e.g. near the biological cell membranes.

7. The Layman's Summary

(describe <u>in layman's language</u> the nature, significance and value of the research project, in no more than 200 words)

We fabricated various types of monodispersed colloidal ellipsoids and measured their motions and forces near or on the liquid-air interfaces. We found novel equilibrium positions near the interface and four types of motions for particles near the interface. We studied dense monolayers of ellipsoids as model systems of glasses and showed that the liquid-to-glass transition exhibits critical behaviors which answers the basic question about the nature of glass transition. It is the first discovery of critical behavior in the rotational motions and two separate critical behaviors during the glass transition. We also discovered glassy dynamics in a buckled colloidal crystal, which can solve the puzzle observed in molecular crystals. We identified the boundary between fine-grained polycrystal and glass for the first time, which bridges the two fields. We also performed the first experiment about the glass surfaces with single-particle dynamics and discovered two surface layers. These results are of fundament importance to glass transition.

We developed novel experiment techniques for the accurate measurements of particles motions and forces near or on the interfaces, which enable to achieve novel experimental results. Moreover, these techniques can be applied to many other studies in the future.

<u>Part C: Research Output</u>

8. Peer-reviewed journal publication(s) arising <u>directly</u> from this research project

(Please attach a copy of each publication and/or the letter of acceptance if not yet submitted in the previous progress report(s). All listed publications must acknowledge RGC's funding support by quoting the specific grant reference.)

The Latest Status of Publications				Author(s) (***bold** the authors belonging to the project teams and denote the corresponding author with an asterisk**)	Title and Journal/ Book *(with the volume, pages and other necessary publishing details specified)*	Submitted to RGC *(indicate the year ending of the relevant progress report)*	Attached to this report *(Yes or No)*	Acknowledged the support of this Joint Research Scheme *(Yes or No)*	Accessible from the institutional repository *(Yes or No)*
Year of publication	Year of Acceptance *(For paper accepted but not yet published)*	Under Review	Under Preparation *(optional)*						
2016				**Bo Li**, **Di Zhou** and **Yilong Han***	Assembly and phase transitions of colloidal crystals (invited review), *Nature Reviews Materials*, 1, 15011	2019	Yes	Yes	Yes
2016				**Feng Wang, Di Zhou** and **Yilong Han***	Melting of colloidal crystals (invited review), *Adv. Funct. Mater.* 26, 8903–8919	2019	Yes	Yes	Yes
2016				**Dongshi Guan**, Yong Jian Wang, Elisabeth Charlaix, and **Penger Tong***	Simultaneous observation of asymmetric speed-dependent capillary force hysteresis and slow relaxation of a suddenly stopped moving contact line, *Physical Review E*, 94, 042802	2019	Yes	Yes	Yes

2017				**Dongshi Guan**, Chloé Barraud, Elisabeth Charlaix, and **Penger Tong***	Noncontact Viscoelastic Measurement of Polymer Thin Films in a Liquid Medium Using Long-Needle Atomic Force Microscopy, *Langmuir*, 33, 1385-1390	2019	Yes	Yes	Yes
2017				Yusheng Shen**,** **Dongshi Guan**, Daniela Serien, Shoji Takeuchi, **Penger Tong**,* Levent Yobas,* and Pingbo Huang*	Mechanical Characterization of Microengineered Epithelial Cysts by Using Atomic Force Microscopy, *Biophysical Journal*, 112, 398–409	2019	Yes	Yes	Yes
2017				**Y. Su**, X.-G. Ma, P.-Y. Lai and **P. Tong***	Colloidal diffusion over a quenched two-dimensional random potential, Soft Matter 13, 4773	2019	Yes	Yes	Yes
2017				**Di Zhou, Feng Wang, Bo Li**, Xiaojie Lou, and **Yilong Han***	Glassy Spin Dynamics in Geometrically Frustrated Buckled Colloidal Crystals, *Physical Review X*, 7, 021030	2019	Yes	Yes	Yes

2017				**Xin Cao, Huijun Zhang** and **Yilong Han***	Release of free-volume bubbles by cooperative-rearrangement regions during the deposition growth of a colloidal glass, *Nature Communications*, 8, 362	2019	Yes	Yes	Yes
2017				**Yun Su**, Pik-Yin Lai, Bruce J. Ackerson, **Xin Cao, Yilong Han, Penger Tong***	Colloidal diffusion over a quasicrystal line-patterned surface, *J. Chem. Phys.* 146, 214903	2019	Yes	Yes	Yes
2018				X. He, **Y. Wang**, and **P. Tong***	Dynamic heterogeneity and conditional statistics of non-Gaussian temperature fluctuations in turbulent thermal convection, Phys. Rev. Fluids 3, 052401(R) (2018).	2020	Yes	Yes	Yes
2018				**Huijun Zhang** and **Yilong Han***,	Compression-induced polycrystal-glass transition in binary crystals, Phys. Rev. X, 8, 041023	2019	Yes	Yes	Yes

2018				**Y. Wang, W. Xu**, X.-Z. He, H.-F. Yik, X.-P. Wang, J. Schumacher and **P. Tong***	Boundary layer fluctuations in turbulent Rayleigh-Bénard onvection, J. Fluid Mech. 840, 408	2019	Yes	Yes	Yes
			2020	**Zhongyu Zheng**, Ran Ni, Hanhai Li, Marjolein Dijkstra, Yuren Wang* and **Yilong Han***	Translational and RotationalCritical-like Behaviors in glass-forming colloidal ellipsoid monolayers	2019	Yes	Yes	No

9. Recognized international conference(s) in which paper(s) related to this research project was/were delivered *(Please attach a copy of each delivered paper. All listed papers must acknowledge RGC's funding support by quoting the specific grant reference.)*

Month/Year / Place	Title	Conference Name	Submitted to RGC *(indicate the year ending of the relevant progress report)*	Attached to this report *(Yes or No)*	Acknowledged the support of this Joint Research Scheme *(Yes or No)*	Accessible from the institutional repository *(Yes or No)*
April/2015/ Hong Kong	Dynamics of Soft Matter at a Three-phase Contact Line	Physics Colloquium, Department of Physics, Chinese University of Hong Kong	2019	Yes	Yes	No
January/2016/Hong Kong	Glassy spin dynamics in buckled colloidal crystal	CityU-PKU Joint Workshop on Disorder and Disordered Materials	2019	Yes	Yes	No
January/2016/HKUST, Hong Kong	Colloidal transport and dynamics over periodic potentials	IAS Computational and Mathematical Problems in Materials Science	2019	Yes	Yes	No
October/2016/Xiamen, China	Assembly of 2D glasses by crystal-to-glass transition and by vapor deposition	Dutch-China Soft Matter Workshop	2019	Yes	Yes	No

March/2017 /Xiamen, China	Interfacial pinning, hysteresis and dynamics of a moving contact line	the 10th National Conference on Soft Matter and Biophysics	2019	Yes	Yes	No
March/2017 /Osaka, Japan	Making glass from crystal and vapor	International workshop on glasses and related nonequilibrium systems	2019	Yes	Yes	No
June/2017/Shanghai, China	Compressing crystal to glass	3rd Conference on Condensed Matter Physics	2019	Yes	Yes	No
July/2017/Xi'An, China	Glassy dynamics in frustrated spin systems	4th National Statistical Physics Conference	2019	Yes	Yes	No
Aug./2017/ Beijing, China	Making glasses from vapor and crystal	KITS Workshop: From supercooled liquids to glasses	2019	Yes	Yes	No
Jan./2018/Hong Kong	Critical-like behaviors and glass transitions in dense monolayers of colloidal ellipsoids	Physics of Supercooled Liquids Workshop at IAS of CityU	2019	Yes	Yes	No
Apr./2018/ Singapore	Compressing binary crystals into glasses	Designer Soft Matter Workshop,	2019	Yes	Yes	No
Nov./2018/ Xiamen, China	Compressing binary crystals into glasses	10th Dyanmics Days Asia Pacific (DDAP10)	2019	Yes	Yes	No
Nov./2018/ Chongqing, China	Compression induced crystal-to-glass transition	11th Conference of Soft Matter and Biophysics	2019	Yes	Yes	No
June/2017/ Montpellier/ France	Interfacial pinning, hysteresis and dynamics of a moving contact line	Soft Matter Seminar, Laboratory Charles Coulomb, University of Montpellier	2019	Yes	Yes	No
May/2018/ Schloss Ringberg/ Germany	Interfacial pinning, hysteresis and dynamics of a moving contact line	Max Planck Workshop on Soft Matter at Interfaces	2019	Yes	Yes	No
October/2018/Bordeaux/ France	Interfacial pinning, hysteresis and dynamics of a moving contact line	International Conference on Liquids at Interfaces in honor of Prof. Elisabeth Charlaix,	2019	Yes	Yes	No

January/2019/ Hsinchu/ Taiwan,	Colloidal diffusion over complex potential landscapes: From periodic, quasi-periodic and random potentials to live cell membranes	2019 Winter School on Frontiers of Complex Systems Science: Soft matter, biophysics and statistical physics, National Center for Theoretical Science	2020	Yes	Yes	No

10. Student(s) trained *(Please attach a copy of the title page of the thesis.)*

Name	Degree registered for	Date of registration	Date of thesis submission/ graduation
Runzhang XU	M. Phil in Physics	Sept 1, 2014	Aug, 2016
Yun SU	Ph.D. in Physics	Sept 1, 2011	May 31, 2017
Dongshi GUAN	Ph.D. in Physics	Sept 1, 2010	Mar, 2016
Yin Wang	PhD in Physics	Sept 1, 2011	Dec 19, 2017
Chengjie Luo	MPhil in Physics	Sept 1, 2016	Aug 31, 2018
Di ZHOU	M.Phil. in Physics	Sept 1, 2014	Mar. 2017
Hongyang TANG	Joint Ph.D. in Physics at HKUST (Hong Kong) and Univ. of Montpellier (France)	Sept.1, 2015	Expected Aug. 2020
Bo LI	Ph.D. in Physics	Sept.1, 2012	Oct. 2017
Xin CAO	Ph.D. in Physics	Sept.1, 2012	Mar. 2017

11. Other impact *(e.g. award of patents or prizes, collaboration with other research institutions, technology transfer, etc.)*

The PIs and co-PI are experimentalists, thus we collaborated with the theoretical groups in other research institutions, including Prof. Pik-Yin Lai of National Central University in Taiwan, Prof. Bruce Ackerson of the Oklahoma State University in the US, Prof. Elisabeth Charlaix of University of Grenoble in France, and and Prof. Jörg Schumacher of Technische Universität Ilmenau in Germany, and the experimental group of Prof. Yuren Wang and Prof. Zhongyu Zheng in Institute of Mechanics, Chinese Academy of Science.