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## PHYSICS OF COLLOIDS IN SPACE-2 (PCS-2)

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## Abstract

Physics of Colloids-2 (PCS-2) The experiment is aimed at investigating the basic physical properties of several types of colloidal suspensions. The three broad classes of colloidal systems of interest are binary colloids, colloid-polymer mixtures, and fractal gels. The objective is to understand their phase behavior as well as the kinetics of the phase transitions in the absence of gravity. The nucleation, growth, and morphology characteristics of the crystals and gels that form would be studied using confocal microscopy. These will be observed directly with excellent time resolution, and therefore extensive information about the different phases and their growth mechanisms will be gained. With the laser tweezers, it will be possible to measure the strength of these structures and to modify them in a controlled way, and the spectrophotometer will provide the possibility to probe their optical properties. We believe that this experiment will provide the basis for future "colloid engineering" in which complicated structures with novel properties (e.g. photonic crystals) will be grown by controlled self-assembly.

## Introduction

Colloidal systems are fluids with other particles dispersed in them, especially particles of size one nanometer to one micrometer. Colloidal systems are found everywhere in nature and in industrial processes. Aerosols, foam, paints, pigments, cosmetics, milk, salad dressings, and biological cells are examples of colloidal dispersions or suspensions. In addition, the

Copyright © 2001 by the American Institute of Aeronautics and Astronautics, Inc. No copyright is asserted in the United States under Title 17, U. S. Code. The U. S. Government has a royalty-free license to exercise all rights under the copyright claimed herein for Government Purposes. All other rights are reserved by the copyright owner. colloidal particles can serve as model systems for the study of fluid and solid properties since they can be considered as playing the role of atoms; hence, they are of interest in the study of the nature of, and transitions among gaseous, liquid, solid/crystal, and glass states of matter. The sizes, shapes (spherical, rods, etc.), and the volume fractions of the particles involved, the surface charge types and distributions, the properties of the fluid medium, and hence the resulting interactions among the particles that can be finely tuned to vary from highly repulsive, to weakly attractive, to strongly attractive interactions, offer a very wide field of colloids research.

The objective of this PCS-2 experiment is to study colloidal systems with particles that are of different size and materials to produce new useful materials. In microgravity, particles of different densities do not separate away by sedimentation. Hence, in space, crystals can be grown with two different kinds of particles, which cannot be done in earth gravity. Thus, for example, large crystals containing both metals and plastics can be grown in microgravity. Such crystals have interesting optical properties such as photonic band gap. By such an approach of 'colloid engineering', we can manufacture a large variety of crystals that are valuable in diverse technical applications.

This experiment is one of the first four experiments that would be conducted in the LMM (Light Microscopy Module)<sup>1,2</sup> in the FIR (Fluids Integrated Rack)<sup>3</sup> in the ISS (International Space Station). The ISS provides long durations of lowgravity environment, and the LMM is a reusable experiment platform that provides confocal- and videomicroscopy, laser tweezers, and spectrophotometry, as well as the possibility to study hundreds of samples. Thus, for the first time, a low-gravity environment is available that suits the needs of long-term experiments that rely on advanced direct microscopic imaging techniques. The colloidal systems that would be studied in the PCS-2 experiment, the need for microgravity, some related ground and flight experiment work, the hardware and flight project development work at NASA GRC, and the proposed experimental procedure are described below.

## Science objectives

The PCS-2 experiment will focus on three classes of colloidal samples, namely, Binary Colloidal Crystals, Colloid-Polymer Mixtures (Gels/Crystals), and Fractal Aggregates and Gels. Quantitative data on nucleation, growth, coarsening, morphology/ structures, and mapping of phase diagrams will be obtained by studying the 'local' structures. that form via self-assembly, in the absence of gravity. These three classes are discussed further below. The actual colloid samples that would be investigated are discussed in the samples selection section.

## Binary Colloidal Crystal Alloy

It is known that under appropriate conditions, monodisperse colloidal particles can self-assemble into crystalline structures with long range periodic order<sup>4</sup>, driven solely by entropy. If particles of different diameters are mixed together, these same entropic effects can lead to the self-assembly of binary alloy crystals<sup>5,6</sup>.

Under certain conditions, it has been found that "hard-sphere" particles (colloidal PMMA) at size ratio 0.58 formed both the  $AB_2$  and the  $AB_{13}$ superlattice structures. Several different crystalline structures have been observed, and more are predicted to occur<sup>7</sup>. In PCS-2, we will use colloidal particles of different materials to make binary alloy crystals. We will study the phase behavior and crystallization properties of these binary alloys.

The phase diagram of binary colloidal crystal alloys, their growth kinetics, the morphology of the crystals formed, and the elastic (rheological) properties of these crystals are of interest.

## Colloid-Polymer Mixtures (Gels/Crystals)

The second set of samples will be a mixture of colloidal particles with polymers. The addition of the polymer induces weak attractive interactions between the colloidal particles by the depletion mechanism, leading to a rich phase behavior for the colloidal particles.

Several such systems, including emulsion droplets<sup>8</sup>, charge-stabilized polystyrene spheres<sup>9</sup>, and polymethylmethacrylate (PMMA) particles<sup>10</sup> have been studied. As the strength of the attractive interaction is

increased by increasing the polymer concentration, the fluid-solid coexistence extends over an increasing range of colloid concentrations. However, the approach to the ultimate equilibrium structure becomes obscured by the kinetics of the phase behavior.

The focus will be on the behavior near coexistence regions, where two or three phases exist simultaneously in the sample. We will study the structure and behavior of these phases, addressing critical questions that have been obscured in previous studies because of limitations imposed bv sedimentation. For example, we will study the nature of the fluid droplets that form and will attempt to measure their interfacial tension by measuring their shape fluctuations. We will study the crystallization of the solid phase, measuring both the structure and the morphology of the crystallites. We will investigate whether the crystals are formed initially in the fluid droplets or whether they can sublime from the gas phase. We will also investigate the potential of using this weak attractive interaction as an alternate means for controlling the growth of colloidal crystals for use as new materials. Finally, we will use the depletion interaction to induce gelation and will study the properties and aging of the resultant gels.

The growth properties of colloidal gels, the nature of equilibrium structure (crystal or gel), the structure and morphology of colloidal crystals, and the elastic (Rheological) properties of gels are of interest.

## Fractal Gels

In the above Col-Pol mixtures, as the polymer concentration is increased, the strength of the attractive interaction becomes so large that the colloidal particles form a gel-like structure<sup>11</sup>. This is characterized by a fractal structure at short length scales and a liquid-like ordering at larger length scales, resulting in a ring of intense light scattering at low angles. This structure is similar to that observed in irreversible aggregation and gelation. Ultimately, this gel-like structure should anneal into a crystalline order; however, under normal gravity the gel can not support its own weight and ultimately collapses, leading to macroscopic phase separation, obscuring the true equilibrium behavior.

Using the confocal microscope, it is possible to image the complete structure directly. Moreover, it is possible to follow the thermal motion of the particles in the gel, allowing the excitation spectrum to be determined. This can be directly related to the elastic modulus of the network; it can also provide a direct probe of the vibration modes of the networks. In addition, we will investigate the slow aging of the structure, and compare these to current theories on aging. Thus, the nature of the polymer gels at "short" length scales, the growth properties of colloidal gels, the largest fractal scales of materials as to how large the scaling properties extend, and the elastic (rheological) properties of colloidal gels are of interest.

Colloid Engineering and Applications:

The use of mixtures of two different materials greatly increases the flexibility of the resultant structures; for example, the characteristic length scale of the structure can be set by one material, which could be an inert plastic, while the second material could have some completely different property, and could, for example, be an optically active semiconductor particle. This provides an opportunity to synthesize structures that are ordered on the length scale of light in all three dimensions, and such materials should have fascinating new properties.

Abundant biomedical applications for colloidal crystals are being developed: drug delivery, biomimetic assemblies, encapsulating cells, tissue culture, controlled release of drugs, flavors, nutrients, and fragrances, and so on. Also, the experimental methods and approaches of these studies, such as the confocal microscopy, laser tweezer, spectrophotometry, and the various video microscopy methods are highly applicable to the development of biomedical sciences.

For the nano/info technology, the material properties of interest are the various optical properties that lead to optical switches, optical filtering, wave guides, and simultaneous three dimensional diffraction. The photonic band gap crystals are patterned with a periodicity in dielectric constant, which can create a range of 'forbidden' frequencies, called the photonic bandgap. This provides an opportunity to shape and mold the flow of light for photonic information technology<sup>12,13</sup>.

One of the most interesting features of photonic band gap structures is their influence on emitters embedded within the crystal. Because the phase space is restricted, the emission properties will be dramatically modified; both the lifetime and the frequency of the emission should be changed<sup>14</sup>. Such effects of modification in the optical properties would be pronounced in colloid based materials also. In addition, the behavior of localized defects in the structure will be analogous to dopants in traditional crystals, and will introduce new optical properties<sup>15</sup>. In order to create these localized structures, we will manipulate the individual particles that make up the binary alloy using laser tweezers. For example, once the binary crystals are formed, a class of experiment that we plan to perform is the local modification of their structure through the manipulation of individual particles using laser tweezers.

## Need for microgravity:

The formation of colloidal crystals is strongly affected by sedimentation; this is most graphically demonstrated by the results of the experiments of Chaikin and Russel, who showed that the morphology of colloidal crystals grown in space is completely different from that grown on earth.

As the crystals sediment, the shear of the fluid flowing past their edges is sufficient to destroy them. In addition, the sedimentation time of the crystals rapidly begins to compete with the diffusion time of the accreting particles, significantly changing the growth mechanism. While some of this effect can be mitigated by buoyancy matching, this is not completely effective, even at the best level of buoyancy match that can be achieved. By calculating the effective Peclet number, (the ratio of the diffusion time scale of the particles, to the settling time scale of the crystal,) it can be shown that the size of the crystals,  $R_{C,max}$  that can be formed varies inversely as the square root of both g, the gravity, and  $\Delta \rho$ , the residual density difference after density-matching. i.e., if we improve the buoyancy match by two orders of magnitude, the size of the crystals will increase by one order of magnitude; by comparison using the standard nonbuoyancy matched fluids, but doing the experiment in microgravity gains an additional 3 decades; this is consistent with what is seen in the CDOT experiments. Combining the buoyancy match and microgravity will produce crystals of remarkable sizes.

In addition, when colloidal particles of different materials are used (e.g., Gold, ZnS, PMMA, Silica, and various core-shell particles, and hollow particles that can provide the desired variation of dielectric constants to the crystal), it is even more essential to perform the experiments in microgravity; otherwise the differential sedimentation of the different particles will prevent growth of any crystals.

## PCS-2 related ground based work:

Several aspects of the colloids research, applications, and use of diagnostic tools such as the confocal microscopy, related to this experiment are described in the PI's<sup>16</sup>, and the Co-I's<sup>17</sup> web pages. Of particular interest to PCS-2 flight experiment are the developments in the colloidal particles manufacture, the studies on binary alloys, col-pols, fractal gels, and the experience in using the confocal microscope and the associated control and data analysis software. These are briefly discussed below.

Among PHaSE-2, PCS-2, and  $L\Phi CA^{18}$  experiments, there is a strong collaboration with Pusey's<sup>17</sup> and van Blaaderen's<sup>19</sup> labs for related

scientific work, and preparation of several types of colloid samples for these research. Several PhaSE-1, PCS-1 experiments' PMMA particles, and those for the ground based work in these labs, and NASA GRC for the work with LMM came from Prof. Pusey, and Schofield's<sup>20</sup> labs; currently, van Blaaderen is working on certain types of core-shell particles; for example, they have produced 1 micron dia silica particles that have 400 nm fluorescent cores. Such fluorescent coresilica shell particle increase the spatial resolution with which these systems can be studied using confocal microscopy. Similarly, silica particles with a gold core (gold radius 15 nm, total 150 nm) have also been produced<sup>19</sup>. These types of particles can be mixed with simpler silica particles to produce crystals with novel properties.

Also of interest are polystyrene spheres coated with ZnS; polystyrene-ZnS core-shell spheres with a 0.2  $\mu$ m radius of the polystyrene core and approximately 60 nm (0.06  $\mu$ m) of ZnS shell thickness have been manufactured. <sup>21</sup>

In addition, at Edinburgh there is an active research effort investigating the phase behavior of mixtures of colloids and polymers. Light scattering studies, that give averaged properties of the various phases that develop, are carried out. Also, direct visualization of the colloidal particles using DIC microscopy is developed; these give detailed local information on the various phases involved as they develop.

At Harvard, an important development that Segre had undertaken is to find a fluid, cycloheptyl bromide, that, when mixed with decalin, allows the PMMA particles to be both index matched and nearly buoyancy matched. With this fluid, it is possible to improve the buoyancy match of the particles by close to two decades in density difference,  $\Delta \rho$ , (from 0.25 to 0.002). This work on dynamics of the crystallization of binary alloys using buoyancy matched mixtures is to complement the work of Schofield in Edinburgh.

Also, experiments to probe growth and dynamics of colloidal gels formed by colloid-polymer mixtures are conducted. This is being done to compare with the behavior of gels formed by irreversible aggregation of colloidal particles. We have found that gels can form at much lower volume fractions using the near buoyancy match solvent, but these gels have a very unusual behavior. We show that gelation of weakly attractive colloids is remarkably similar to the colloidal glass transition. Like the glass transition, dynamic light scattering functions near gelation scale with scattering vector, and exhibits a two-step decay with a power-law divergence of the final decay time. Like the glass transition, static light scattering does not change upon gelation. These results suggest that, like the glass transition, gelation results from kinetic arrest due to crowding of clusters, and that both gelation and the glass transition are manifestations of a more general jamming transition<sup>22</sup>.

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In addition, extensive work in being carried out to develop the use of confocal microscopy to measure structure and dynamics of colloidal systems in 3D. Three dimensional colloidal crystallization were observed as follows. Laser scanning confocal microscope from Noran Instruments was used to image 3D regions in PMMA beads that contain a fluorescent dye. By taking images two or three times every minute crystallites formation and evolution with time was recorded. After the measurement the positions of the particles were determined and tracked.



Here, figure caption

The algorithm for finding crystalline regions relies on the assumption that the neighbors of a particle in a crystal lattice are arranged in a particular orientation about the particle, and that this orientation of neighbors is the same for nearby particles. Particles with many crystal-like 'bonds' (e.g. 8) are said to be 'crystal-like'. Such an algorithm and analysis of the confocal image data give us insight into the crystals as they are just nucleating, and growing with time. Particles with a crystal-like surrounding are represented by the large red spheres while the smaller yellow spheres represent particles in the metastable liquid.

In another work, the nature of glass transition is studied. We used small colloidal particles to model atoms in a glass, and studied them using confocal microscopy. As the particles are packed together, if one of them wants to move, its neighbors have to cooperate. As they are packed even tighter, more of the particles have to cooperate for any to move. When all of the particles in the sample have to cooperate, the sample is essentially a solid -- thus explaining what is happening with glass as it's cooled. This is a classic theory, and for the first time we can look at a real physical experiment and directly see cooperative motion.<sup>23</sup>



#### Here, Figure. caption

The picture shows experimental data of cooperative clusters of particles (the largest in this image is highlighted). All of the particles are actually the same size; the slower particles are shown smaller so that the fast ones stand out.

#### PCS-2 and the related Flight experiments

Several precursor experiments supporting the goals of PCS1<sup>24,25</sup>, and PCS-2 have been flown as glovebox experiments aboard the Russian space station MIR. These helped provide critical tests to determine the effects of microgravity on colloidal dispersions. Four glovebox experiments (BCAT-1, BCAT-2, CGEL-1, CGEL-2) have been performed to date.

BCAT-I successfully produced binary alloy crystals of the AB<sub>13</sub> structure. Samples were chosen near the optimal size ratio of 0.58, and at several different total volume fractions. Interestingly, the optimum volume fraction for the crystal growth turned out to be 0.54; this was higher than the optimum value on earth. This highlights the difference between results obtained on earth and those obtained in microgravity; this result was also extremely useful in planning for PCS-1. The results of BCAT-I also included the first observation of the persistence of colloidal crystals formed from monodisperse emulsion particles. On earth, these emulsions cream and, even though crystals do form, they do not persist when the emulsion creams. These results suggest that this may

be due to the creaming; as the crystallites increase in fraction, they become unstable to volume rearrangements. This may be due to the intrinsic instability expected for an FCC (or RHCP) lattice of particles with liquid films at their interfaces; these films must then meet the Plateau criteria for stability which can not be done for an FCC structure<sup>26</sup> Finally. the video component of BCAT-I also proved conclusively that the instability of the kinetically arrested gel-like structure formed upon the addition of high concentrations of polymer to induce a very strong depletion attraction among the PMMA spheres is gravity-induced. All earth experiments have shown that this gel structure collapses after some delay time, which depends on the strength of the attraction. Such a collapse precludes investigation of the long term stable state of the sample, which is completely unknown. The collapse was not observed in microgravity, but there was insufficient time to monitor the evolution of the long term structure.

BCAT-II tested a different series of binary alloy mixtures. A goal here was to further explore the phase boundaries of the region where good colloidal crystals form as r is varied. The samples chosen for BCAT-II had r=0.61, which is slightly to the high side of the optimum value determined on earth. The goal was to investigate whether the optimum value of r changes as does that of  $\phi$ . The results suggested that this size ratio is very inefficient in forming colloidal crystals, implying that the optimum value of r is the same in microgravity as on earth, unlike the value for  $\phi$ .

CGel tests were designed to perform light scattering studies of all three classes of samples to be flown on PCS-1, including the binary alloys, the colloid-polymer mixtures and the fractal colloidal aggregate gels. The pictures from these tests suggest that the crystal phase forms within the fluid phase, and does not sublime from the solid phase. However, the magnification of the camera lens used for these photographs was not sufficient to confirm this unambiguously, and further experiments to determine this will have to await PCS-2.

In the CGEL-2 tests, on the John Glenn shuttle mission, we were able to form an  $AB_6$  colloidal crystal, a structure that was discovered in Edinburgh only a few months before the mission. This sample grows quite rapidly as compared to most other binary allows, allowing us to study the crystals grown in space. Interestingly, no dendritic growth of the large crystallites was seen, suggesting that the growth mechanism differs significantly from that of monodisperse particles. We were also able to form colloidal fractal aggregate gels, both from irreversible aggregation and through the use of the depletion attraction.

The PCS-1 experiment is currently being conducted in the ISS, in the EXPRESS rack. In PCS- $1^{24}$ , three classes of colloidal materials are being studied. These include ordered crystalline samples; mixtures of colloidal particles with other species, primarily polymers, which induce a weak attractive interaction allowing us to precisely tune the phase behavior of the mixtures; and highly disordered, but very tenuous fractal structures which possess their own unique symmetries and their own unique properties. The PCS-1 experiment will yield considerable information about the bulk properties of the colloidal dispersions.

In PCS-2, we will emphasize materials synthesis, and will extend the range of materials used. In addition, we will study the structure and dynamics of these systems in real space, allowing us to probe local structure in unprecedented detail.

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## Flight Hardware and Development work at NASA GRC

To accommodate a larger set of flight experiments, a Leica RXA microscope is modified to accommodate various video microscopy techniques, interferometry, confocal microscopy, laser tweezing, and spectrophotometry. The imaging techniques of high resolution color video microscopy, bright field, dark field, phase contrast, differential interference contrast (DIC), fluorescence, spectrophotometry, and confocal microscopy are combined in a single configuration. An experimenter can choose from six (6) objective lenses of different magnifications to achieve the required science data. This suite of measurements allows a very broad characterization of fluids, colloids, and two-phase media, including biological samples.<sup>27</sup>.

To translate the 1-g experiments into microgravity, there are several required repackaging, and design exercises; these are taken care of at NASA GRC. Tele-metry and tele-commanding aspects require development efforts for automation, software, and training. Also, vibration, temperature cycling, EMI tests, vacuum/fluid-leak, acoustic noise emission, and safety are some of the other standard issues that are important.

Additionally, several experiment specific important issues arise; there has been excellent communication between the engineering and the research teams, regarding such issues that can affect the science and engineering, and various new ideas, and/or compromises are continually developed. Some such issues are solved or compromised, either at NASA GRC or at the PI labs. Some such sample issues are listed below:

The ability to contain the fluid samples for long duration, without leaking is tested, and various types of sample-cell developments were carried-out and evaluated. Various methods of sample homogenization techniques were tested and evaluated; shelf-life and compatibility of the sample themselves are important; the type of lasers used need to be compatible with the dyes used; and the laser light has a bleaching effect on the fluorescent samples, and hence on the life of fluorescent dyed particles; various chunks of experimental work need to be time-lined within the microgravity, and crew- time constrains -- all such issues demand a healthy interaction among the PCS2 team members, and suitable solutions were found; such matters were/are reviewed in detail in the various NASA reviews such as the (RDR) Requirement Definition review) and PDR (Preliminary Design Review) and so on.

Also, to avoid overdesign, several questions come up and appropriate compromises or solutions were found. Some issues of this nature are: we do not need the ability to visualize the tweezer beam since it can be worked out; for spectrophotometer we can relax the polarized light input requirement, for cells that could use application of electric field, the field can be applied on all cells in the slide rather than requiring individual electrical connections; All such solutions and/or compromises save development time and cost.

In the interest of cost-saving and yet keeping the value for broad science, including biological sciences, one of the most important compromise that was undertaken was the inclusion of confocal microscopy, while relaxing the requirement for laser light scattering techniques that are used in the precursor colloids experiments. The design of the microscope still retained the ability to add new capabilities that can become desirable in future. Such a decision was made at an early stage of the project and it continues to prove to be an excellent decision. The details and the capabilities of the LMM itself are described in the several documents<sup>27-29</sup>.

## Experimental procedure

Binary Alloys: Once the sample is homogenized, the nucleation and growth of individual crystallites of the binary colloidal alloy will be monitored with the confocal microscope, by taking three dimensional stacks of images. Confocal microscopy will allow to make observations about the shape and the structure of the nuclei that are not possible with the light scattering techniques used in PCS-1, since not only the average structure of the nuclei but also its variations within a nucleus can be studied. The comparison with analogous ground-based experiments will reveal differences in the growth behavior under micro-gravity. Series of two dimensional pictures taken in fast succession will allow to track all visible particles. This will be done either by video microscopy or with the confocal. It allows to characterize the way how particles move and diffuse in the sample.

Once large crystallites have formed we will investigate the properties of the crystal lattice. Again the structure of the stable structure can be determined by taking 3D stacks of images with the confocal. In order to measure the elastic properties, a test bead within a crystallite can be grabbed with laser tweezers and be used to distort the lattice. The bead is vibrated back and forth with the laser tweezers in order to create collective excitations (the equivalent of phonons) that are then observed by video- or confocal microscopy. Micro rheology will be used to find the elastic properties in the linear regime. In order to do this the motion of a particular bead will be tracked. This can be done by either confocal or video microscopy.

The laser tweezers will also be used to modify the crystal lattice by creating defects.

<u>Colloid-Polymer Mixtures</u>: The colloidal particles for this experiment will again be dyed PMMA spheres suspended in an index matched fluid. The volume fractions of these samples will range from about 1% to 30%. The polymer will be polystyrene, with a molecular weight ranging from about 100,000 to 1,000,000.

The colloid-polymer mixtures will be homogenized using the same procedure used for the binary colloid mixtures. After they are homogenized, the formation of the gel will be monitored by taking 3D stacks of images in the same way as for the binary colloidal crystals.

During the equilibration of the samples, DIC or phase contrast microscopy (and occasionally also confocal microscopy) will be used to find out which samples will eventually contain gas, liquid or solid or a combination of these. An automatic recognition of the different phases could save a lot of time. This information will be used to establish the phase diagram of this system under micro-gravity, and it will be the input for selecting a set of representative samples that will be studied in more detail. Also this will complete the insight gained from PCS-1 into the mechanisms that lead to the ultimate equilibration of the gel. The selected samples will be observed in longer confocal or video-microscopy runs in order to characterize the present structures and the different phases. Colloidal crystals may form in one of the phases. Since they can form at low volume fractions and result from an attractive interaction between the particles, their growth and evolution may be significantly different from the formation of those formed by hard spheres. Therefore, we will use confocal microscopy to study their

nucleation and growth in a way that is analogous to the one described in the section above about binary colloidal crystals.

In addition, some samples will contain test beads that will be used for micro rheology measurements, and the elastic properties of the gels and solid structures will be measured with laser tweezers (rheology).

<u>Colloid-Metal, Col.-Semiconductor, and Col.-</u> <u>Liquid-Crystal Mixtures:</u> The phase diagrams of samples made of colloid-metal, colloid-semiconductor, and colloid-liquid-crystal mixtures are expected to be similar to the case of binary PMMA mixtures. However, this is difficult to test in ground based experiments, since these samples cannot be density matched by choosing a suitable solvent. Therefore, it will be part of the experiments planned for PCS-2 to map out the details of the phase diagrams of the different sample types.

The experiments needed to study the formation and the evolution of the structures in these samples will be analogous to the ones described in the sections on binary alloy colloidal crystals and colloid-polymer mixtures above. In addition the effect of dc- and acelectric fields on the optical properties of these samples will be determined. In order to do this a sample will be illuminated with monochromatic light under various angles during a video microscopy run.

#### Test Sequences

### HERE, 1 sample...

## Samples selection for pcs-2:

Several ground-based experiments, phasediagram studies, and results from other experiments in micro-gravity will used to guide the selection of the samples that are the most scientifically interesting and the most promising for applications.

A selection list of colloidal samples for these experiments are available if the flight samples were needed now; however, the exact sizes, materials, volume fractions ranges of interest, and their characterization will be continually fine tuned further, in a way that would not affect the hardware development, but rather improve the eventual science return. Currently data from the PCS-1 flight experiment that is currently underway, will also be considered. Binary alloys of PMMA-PMMA, or Silica-Silica, and col-pol systems of PMMApolystyrene can be made available now; currently, more advantageous core-shell particles are being developed and evaluated as described earlier.

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## Summary

The PCS-2 experiment will investigate the nucleation, growth, and morphology of binary alloys. colloid-polymer and fractal gels using video and confocal microscopy. These structures are formed via self-assembly of nano-scale colloidal precursor particles. Laser tweezers will be used to study the rheological properties of the structures and to introduce defects. The optical characteristics will be studied using spectrophotometry. The multi-user light microscope module (LMM) hardware developed is an important and very fruitful hardware that will benefit not only the science of colloids, and condensed matter physics, but also several other important disciplines such as biological sciences, and manufacture and application of novel materials that are relevant to optical and information/computer technologies.

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## References

<sup>1</sup>NASA Glenn Research Center., "Light Microscopy Module (LMM)," http://microgravity.grc.nasa.gov/ 6712/ lmm.html.

<sup>2</sup>Logicon/Northrop Grumman Co., "Light Microscopy Module (LMM)," http://cleveland.feddata.com/LMM/.

<sup>3</sup>Gati, F., and Hill, M., "The FCF Fluids Integrated Rack: Microgravity Fluid Physics Experimentation on Board the ISS," AIAA-2001-4926 (2001).

<sup>4</sup>Pusey, P.N., and van Megen, W., "Phase Behaviour of Concentrated Suspensions of Nearly Hard Colloidal Spheres," *Nature* **320**, (6060), 340-342 (1986).

<sup>5</sup>Eldridge, M.D., Madden, P.A., and Frenkel, D., "Entropy-Driven Formation of a Superlattice in a Hard-Sphere Binary Mixture," *Nature* **365** (6441), 35-37 (1993).

<sup>b</sup>Eldridge, M.D., Madden, P.A., and Frenkel, D., "A Computer-Simulation Investigation into the Stability of the AB2 Superlattice in a Binary Hard-Sphere System," *Mol. Phys.* **80** (4), 987-995 (1993).

<sup>7</sup>Eldridge, M.D., Madden, P.A., Pusey, P.N., and Bartlett, P., "Binary Hard-sphere mixtures: A Comparison between

# Computer Simulation and Experiment," Mol. Phys. 84, 395-420 (1995).

<sup>8</sup>Bibette, J., Roux, D., and Nallet, F., "Depletion interactions and fluid-solid equilibrium in emulsions," Phys. Rev. Lett. 65, 2470-2473 (1990).

<sup>9</sup>Calderon, F.L., Bibette, J., and Biais, J., "Experimental Phase-Diagrams of Polymer and Colloid Mixtures," *Europhys. Lett.* 23 (9), 653-659 (1993).

<sup>10</sup>Ilett, S.M., Orrock, A., Poon, W.C.K., and Pusey, P.N., "Phase-Behavior of a Model Colloid-Polymer Mixture," Phys. Rev. E 51 (2), 1344-1352 (1995).

<sup>11</sup>Poon, W.C.K., Selfe, J.S., Robertson, M.B., Ilett, S.M., Pirie, A.D., and Pusey, P.N., "An experimental study of a model colloid-polymer mixture," *J. Phys.* de Physique II 3 (7), 1075-1086 (1993).

<sup>12</sup>Joannopoulos, J.D., Villeneuve, P.R., and Fan, S., "Photonic crystals: putting a new twist on light.," Nature 386, 143-149 (1997).

<sup>13</sup>Joannopoulos, J. D., "Photonic Crystal Research," http://ab-initio.mit.edu/photons/.

<sup>14</sup>Martorell, J., and Lawandy, N.M., "Spontaneous Emission in a Disordered Dielectric Medium," Phys. Rev. Lett. 66 (7), 887-890 (1991).

<sup>15</sup>John, S., "Strong localization of photons in certain disordered dielectric superlattices," Phys. Rev. Lett. 58 (23), 2486-2489 (1987).

<sup>16</sup>Weitz, D.A., "Experimental Soft Condensed Matter Group, Div. of Engineering and applied sciences/ Dept. of Physics, Harvard University," http://www. deas.harvard.edu/ projects/ weitzlab/.

17Pusey, P.N., "Soft Condensed Matter," http://www.ph.ed.ac.uk/cmatter/cgi-

bin/archive/show.cgi?db=people&id=pusey.

<sup>18</sup>Lin, K.-h., Crocker, J.C., Prasad, V., Schofield, A., Weitz, D.A., Lubensky, T.C., and Yodh, A.G., "Entropically Driven Colloidal Crystallization on Patterned Surfaces," Physical Rev. Letters 85 (8) (2000).

<sup>19</sup>Blaaderen, A.v., "Colloidal Matter,"

http://www.amolf.nl/research/colloidal\_matter/.

<sup>20</sup>Schofield, A., "Andrew's PMMA Latices," http://www.ph.ed.ac.uk/~abs/Latexlist.html.

<sup>21</sup>Dinsmore, A.D., Breen, M.L., Qadri, S.B., and Ratna, B.R., "Photonic Crystals from Core-Shell Colloidal Spheres," (submitted).

<sup>22</sup>Segre, P.N., Prasad, V., Schofield, A.B., and D. A. Weitz, "Glass-like kinetic arrest at the colloidal gelation transition," Physical Review Letters 86, 6042-6045 (2001).

<sup>23</sup>Weeks, E.R., Crocker, J.C., Levitt, A.C., Schofield, A., and Weitz, D.A., "Three-dimensional Direct Imaging of Structural Relaxation Near the Colloidal Glass Transition," Science 287 (627) (2000). <sup>24</sup>Weitz, D.A., Doherty, M., Jankovsky, A., Koudelka, J.M., Ansari, R., Lorik, T., Shiley, W., Bowen, J., Kurta, C., Eggers, J., Bambakidis, K., Hovenac, E., Segre, P., Bailey, A., Gasser, U., Manley, S., Prasad, V., A, S., and Pusey, P., "The Physics of Colloids in Space," Conference & Exhibit on International Space Station Utilization—2001, Cape Canaveral, Florida. 2001-4900 (2001).

<sup>25</sup>Ansari, R.R., Hovenac, E.A., Sankaran, S.,
Koudelka, J.M., Weitz, D.A., Cipelletti, L., and Segre,
P.N., "Physics of Colloids in Space Experiment,"
Space Technology and Applications International
Forum – STAIFF-99, Alburquerque, NM. (1999).

<sup>26</sup>Weaire, D., and Phelan, R., "A Counterexample to Kelvin Conjecture on Minimal-Surfaces," Philosophical Magazine Letters **69** (2), 107-110 (1994).

<sup>27</sup>Motil, S.M., and Snead, J.H., "The Light Microscopy Module: An On-Orbit Multi-User Microscope Facility Planned For The Fluids And Combustion Facility On The International Space Station," Conference & Exhibit on International Space Station Utilization—2001, Cape Canaveral, Florida. AIAA–4956 (2001).

<sup>28</sup>Resnick, A., "Design and construction of a spaceborne optical tweezer apparatus," To appear in Rev. Sci. Instrum. 72, (11) (2002).

<sup>29</sup>Doherty, M.P., Motil, S.M., Snead, J.H., and Malarik, D.C., "Microscope-Based Fluid Physics Experiments in the Fluids and Combustion Facility on ISS," Spacebound 2000, Vancouver, B. C. May (2000).