Please ensure that your submission follows closely the <u>*Guidelines*</u> *for Long Term Projects.*

1. **Abstract:** summarize the strong points of the proposal (in accordance with the *Principles)*

Given the importance of granular matter for industrial processing (about 60% of all processed goods come as granulates) and geological engineering, the origin of its mechanical properties are surprisingly highly unknown. In particular, the liquid distribution in wet granulates, which are responsible for its special mechanical properties and the transport of liquid within granular piles or porous media on the pore level remained obscure. This is mainly due to a lack of experimental techniques. This proposal thus aims to study the interaction between liquids and porous or granular materials and to optimize two techniques recently developed at ID15: ultrafast x-ray tomography and x-ray stereo imaging.

To explore the mechanical properties of wet granulates we propose to track the 3D motion of tracer particles in a strongly fluidized granulate by optimizing a unique x-ray stereo imaging technique with very high time resolution in the millisecond range. Furthermore, we propose to explore the dynamic distribution of liquid in a sheared granulate in real time by capturing movies of ultrafast tomograms. The knowledge of the liquid distribution will be used to study driven liquid fronts in granular piles and porous media with good time resolution. Within the framework of this long term proposal we expect to image driven liquid fronts in real time i.e. with typical driving speeds used for field oil extraction.

To improve the current imaging speed using monochromatic beam we thus propose to replace the Si monochromator with a multilayer monochromator exhibiting an appropriate band width for high energy imaging.

The project combines the expertise of four institutes, having long standing records in the proposed research fields of granular matter, wetting of structured substrates, driven liquid fronts, and x-rays.

We intend to hire up to ten postdoctoral researchers and PhD students to work on these projects. They will work both experimentally and theoretically to develop a complete picture of the dynamics of liquids in fluidized granulates and driven liquid fronts in granulates and porous media.

The new experimental setups and the required new technical developments will enable to image soft condensed matter samples with high speed and a minimum of radiation damage. This is expected to be of value for most of the current users of ultrafast tomography, and will also attract new user groups to the ESRF since it will enable new insights into porous and granular (“soft”) matter.

2. **Scientific Case** and **development aspects** (reasons for requesting a LTP, results expected and scientific/technical goals for scheduling period of the proposed LTP):

Granular matter has been investigated for a long time due to its enormous importance in many fields. More than half of the raw materials world-wide come in granular form. Their transport properties and mixing behavior with liquids thus affects annual money flow on the Tera-dollar scale. Soils, sands, and sedimental rock, including submontane oil reservoirs, are granular systems as well. Furthermore, the prediction and management of land slides or avalanches requires deep knowledge of many aspects of granular physics. In spite of the obvious social, economic, and scientific relevance of granular materials, there is so far only very

limited understanding of the complex mechanisms governing their physical properties. This pertains to dry granular systems, such as sand dunes in the desert or bulk solids in industry, but also wet granulates, or the transport of liquid within granular piles, as it is encountered in oil reservoirs. This (sometimes costly) lack of deep understanding is in part due to the inherent complexity of granular matter, but is also a consequence of the absence of suitable experimental methods which allow time-resolved imaging of grain-scale processes within a granular system. We firmly believe that the ESRF has the potential to provide these experimental means, and we hereby propose their development.

Dynamics of strongly fluidized granulates:

To understand the mechanical behavior of wet granulates in detail it is important to understand the influence of the liquid distribution on the dynamics of a fluidized granulate. In recent years there has been considerable progress in understanding the dynamics of dry granular materials [1]. However, an equally mature understanding of the properties of wet granular materials has not yet been achieved. Studies of particle diffusion in agitated dry granulates yield a variety of results, such as the presence of subdiffusion and transitions from subdiffusive to diffusive flow regimes. The occurrence of these results depends intimately on numerous physical parameters, such as the depth of the flowing layer in the granulate [2] and grain packing density [3]. Since the type of diffusion in dry granular systems depends strongly on physical parameters, it is difficult to predict what type of diffusion is present when grains experience cohesive forces due to the presence of a liquid.

Sheared granulate:

When granulate is sheared, the position of the grains change, and the capillary bridges and larger liquid agglomerates (liquid cluster) break, change their size and shape, or reform. Because liquid bridges need some time to form and grow, the size and number of bridges or clusters, is depending on the shear rate. This results in a shear thinning behavior of wet granulates [4]. In a wide range of liquid content, about six capillary bridges per sphere (coordination number) were found, resulting in a stiff granulate. For larger liquid contents, the liquid forms extended liquid clusters and the stiffness of granulate is expected to be reduced. Whereas in our experiments, the yield stress of the granulates is about constant for a very large range of liquid contents exceeding the regime with coordination number six by far, reaching into the regime where large liquid clusters are present. Hence, the time resolved knowledge of the distribution and shape of liquid bridges and cluster and the corresponding matrix consisting of glass spheres is needed for a quantitative understanding of the mechanical properties of wet granulates. Fast x-ray tomography will provide an adequate and time resolved description of the distributed liquid and the packing of a sheared granulate, which are crucial parameters for the understanding of wet granular material in general. In the proposed experiments, we will explore the dynamic behavior of capillary bridges and liquid clusters within a sheared wet granulate. We will characterize how liquid bridges and clusters change their size and shape as a function of shear rate. This will be done for beads with variable wettability, diameter and, performed as a function of liquid content and shear rate.

Driven liquid fronts in porous media:

The process of (driven) imbibition of a liquid into a random medium plays a key role in many problems of general interest, such as oil recovery, irrigation in agriculture, or wetting of raw material powders in industrial chemistry. Inspired by this tremendous practical importance, many experiments have already been carried out in search of a thorough physical understanding of the basic mechanisms involved. However, a satisfactory agreement with theory has not yet been achieved. This is in part due to difficulties inherent to the experiments performed so far. Well-known problems of imbibition experiments, which use paper as the random medium [5] are swelling, have a poorly defined roughness, and encounter evaporation [6]. Other more conceptual reasons for theories to fail are due to entrainment of air in the advancing front in realistic scenarios. This is captured neither by percolation models nor by theories on interface roughness statistics like the KPZ equation, but may be accounted for by more recently developed phase field models [7]. We could recently show in-situ that interface roughness theory works well for a simple 2D model system [8]. However, results on more realistic 3D systems, like columns of glass beads [9] which allow for a microscopic analysis of the advancing front, exist to date only for ex-situ studies [10]. High resolution real-time mapping of the

advancing front in a realistic setting has never been done up to now, but is indispensable for the development of theoretical models of the temporal evolution of the front topology.

Results achieved so far:

Using the fast x-ray micro-tomography setup at ID15A we could already show that the remarkable insensitivity of the mechanical properties of wet granulate from the liquid content is due to the particular organization of a wetting liquid into open structures in randomly dense packed granulates [11], as shown in Figure 1. We also found a volume exchange process between the individual liquid bridges and clusters with a characteristic time constant of about five minutes that is driven by differences in Laplace pressure [11,12] (Figure 1c). Based on these results on the static distribution of liquid within granular piles and their slow equilibration process we propose to explore fast dynamic processes in wet granular systems.

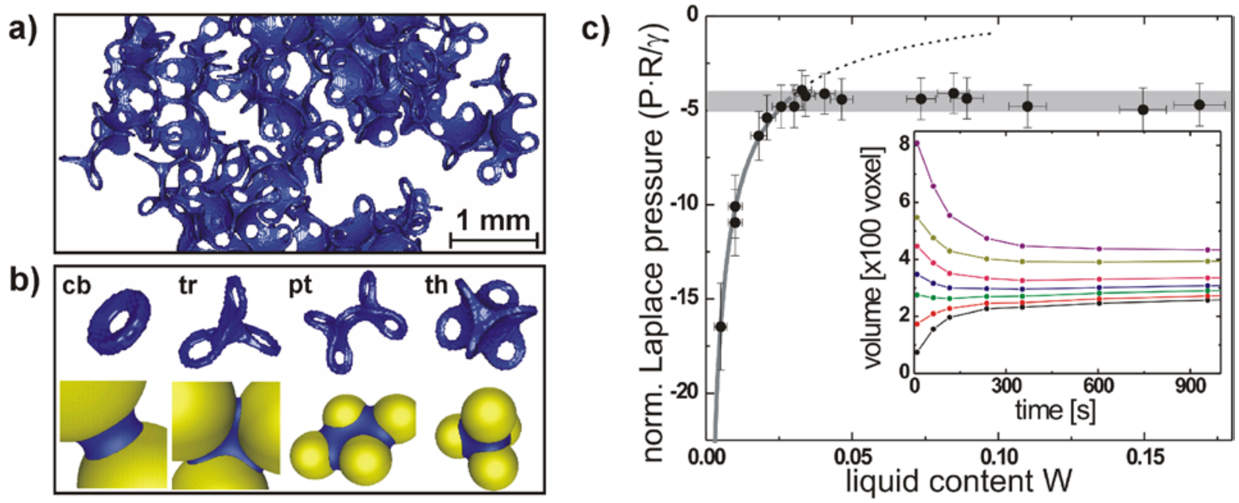

Figure 1: a) Fraction of a large percolated liquid cluster. (a) *Top row*: capillary bridge (**cp**), trimer (**tr**), pentamer (**pt**), and filled tetrahedra (**th**) as found via x-ray tomography. *Bottom row*: as obtained numerically. c) Laplace pressure within liquid clusters, as a function of the liquid content of the sample. The inset shows the equilibration of individual liquid clusters after preparation.

Our previous experiments at ID15A showed that the trajectories of slow tracer particles in fluidized wet granulates can be recorded using fast x-ray imaging. The technique is based on a particle tracking-type imaging protocol: The yz position (plane orthogonal to the x-ray propagation direction) of a few highly absorbing tracer beads are determined by x-ray absorption radiography. Figure 2a shows an absorption image of BaTi tracers undergoing vertical fluidization with surrounding glass spheres, and the tracers' positions can be clearly identified. The corresponding x position (parallel to the beam) is obtained by rotating the sample by about 7 deg, recording a second absorption radiography, as sketched in Figure 2b.

Assuming that the position of the beads did not change between the two images we can calculate the 3D position by comparison of the two consecutive images. Repeating this process continually, we can record 3D motion paths of tracer particles in a fluidized granulate. These preliminary runs have demonstrated that grain speeds of up to 200 µm/s can be recorded with good lateral resolution at ID15A, providing that the oscillation amplitude is not too large. A preliminary calculation of a short flight path is shown in Figure 2c.

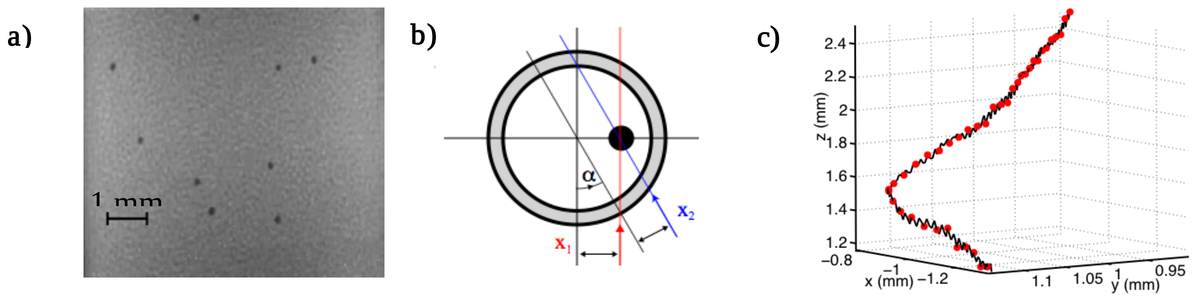

Figure 2: a) typical 2D projection using BaTi tracer beads b) sketch of how to obtain the 3D position of a tracer beads from two 2D projections, taken at different viewpoints c) preliminary reconstructed flight path of a tracer sphere with an average speed of 10 µm/s.

First experiments at ID15A revealed also that a continuously sheared granulate can be imaged with shear rates up to about 0.2 µm/s using the monochromatic beam in 16 bunch mode (Figure 3). With further optimizations of the current setup, it shall be possible to push the shear rate up to 2 µm/s.

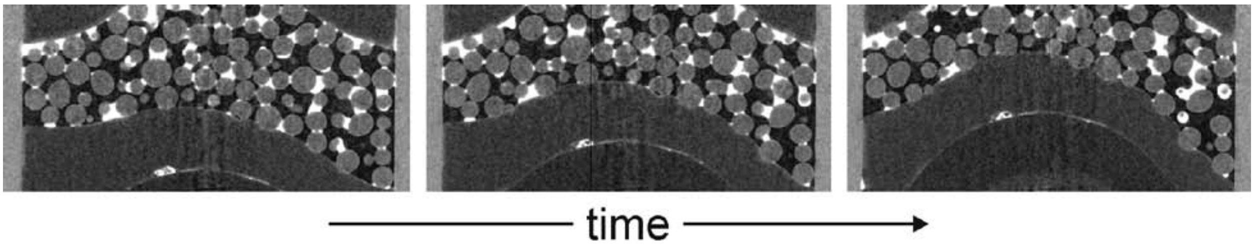

Figure 3 Preliminary results using a home build shear cell showing sections through 3D tomograms of wet granulates consisting of glass beads. As wetting liquid water is used with ZnI_2 solved in it (bright areas).

Furthermore, we have performed test experiments at ID15A in which we could resolve the global shape of a rising liquid front in sintered glass filters with good lateral resolution (Figure 4a). The formation of pockets of trapped air behind the liquid front could be clearly observed. The surrounding glass matrix, the liquid and the air can be unambiguously distinguished from each other. Fast x-ray tomography thus provides an adequate assessment of the structure of the solid matrix as well as the time resolved imbibitions of a liquid into a porous medium. On the pore size level the situation is even more interesting as can be seen in Figure 4b. An area that is once filled with liquid, will not necessarily stay filled as the liquid front propagates. This can be seen comparing e.g. the image at 6 min with the image at 28 or 43 min. A formerly filled area is drained completely once a “hydrodynamic bypass” is formed. At this point it becomes obvious why oil mining companies manage to extract only a fairly small percentage of around 20 – 30 % of the stored oil. We aim at understanding this behavior on a pore size level and to suggest improvements.

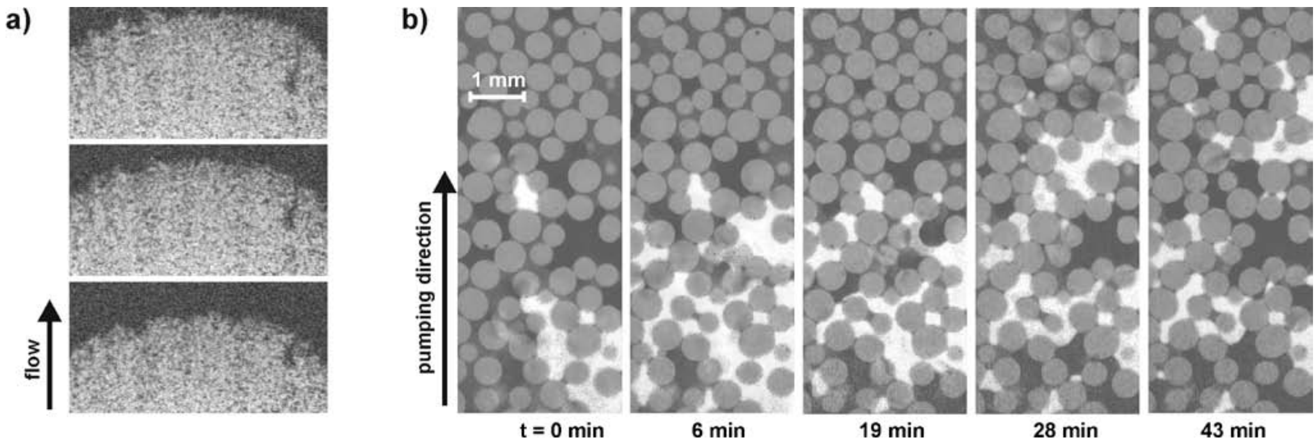

Figure 4: 2-D cross sections of x-ray tomography time series. *a)* Liquid imbibing a sintered glass filter. *b)* Porous matrix consisting of spherical glass beads: driven aqueous front displaces oil.

The ultrafast tomography at ID15A was proven as an ideal tool for research on wet granular matter with much better temporal and lateral resolution that what can be done to date by any other method e.g. Nuclear Magnetic Resonance.

For the experimental setups proposed here, x-ray energies at around 50 keV have best material contrast at minimum sample damage. In fact, using a monochromatic beam no radiation damage was noticeable within continuous exposure of several hours. Hence, to prevent radiation damage to our samples all imaging will be done using a monochromatic beam. Owing to the high temporal resolution available at beam line ID15A, inherent ageing problems in the samples can be circumvented and data of highly absorbing static samples can be obtained in excellent quality. Moreover, the large imaging speed even allows to capture dynamic (re-) organizations of granulates and wetting fluid which will be very important for reaching the goals of our long term proposal. To open new experimental possibilities i.e. the temporal resolution at ID 15A some technical developments are required as we will indicate in the following. To open new

experimental possibilities to a plethora of research topics, some new technical developments are required as we will indicate in the following.

Technical and methodological developments:
To be able to capture also the fast dynamics of strongly fluidized granulate the temporal resolution needs to be improved. For that we propose to replace the Si monochromator with a multilayer monochromator for high energy imaging with appropriate band width. Using the multilayer we expect to gain more than one order in magnitude in x-ray flux.

Dynamics of strongly fluidized granulates:
For the particle tracking, the need to acquire two consecutive images in order to determine the 3D position of a tracer particle obviously limits the maximum detectable velocity (Figure 2). To improve the time resolution by a faster rotation of the sample is limited by the motional smear caused by the rotational velocity that might become larger than the particle velocity one wants to track. To overcome this limitation we propose to further develop a stereo imaging technique based on two intersecting monochromatic x-ray beams in close collaboration with M. Di Michiel. This fast stereo imaging technique will provide a three-dimensional and time-resolved description of the motion path of tracer particles. It will not only improve the time resolution by almost two orders of magnitude (allowing to image tracer speeds up to 10 mm/s using the Sarnoff camera), it will also improve the particle statistics significantly since stereo imaging will help to discriminate the 2D problem of particles "hidden" behind others and thus a larger density of tracer particles can be used. By measuring multiple tracer particle trajectories in a single sample, we can obtain a long time-series of the mean flow profile and velocity field. By subtracting the mean flow from the velocity profile we can obtain the tracer particle displacements [13]. If no large-scale flow pattern is present, we can determine the tracer displacements directly from the flight path. The time dependence of the mean displacement squared of the tracer particles yields the type of diffusion present, while its slope gives the diffusivity. In the proposed experiments, we will investigate how the flow profile and diffusion of the tracers is affected by different oscillation frequencies and amplitudes above the onset of fluidization. This will be performed for grains with variable sizes and liquids.

Sheared granulate:
Shearing wet granulate in a specially designed sample cell we expect to extract the reordering of the granules and the liquid as function of physical parameter. For this we plan to design an experimental setup small enough to fit to the dimensions of the x-ray beam, where granulate can be continuously sheared. We expect to explore effects like e.g. shear thinning, arching, or crack formation. Due to the above mentioned characteristic time scale of the liquid redistribution within a wet granulate it will be very interesting to study not only slow shear rates but also shear rates faster than the liquid redistribution to access e.g. the shear thinning behavior. The found dynamic liquid distribution shall serve as starting point to study driven liquid fronts flowing through a model granulate; i.e. a situation comparably to oil extraction.

Driven liquid fronts:
We will explore the dynamics of driven imbibition of aqueous liquids into a porous medium filled with an organic phase. Particular focus will be on the dynamics and the shape of the liquid front as well as on the complex internal behavior of the fluid inclusions in the solid matrix after the liquid front has passed. We will do this for columns filled with spherical beads and sintered glass filters, with various bead respectively pore sizes as a function of the viscosity and wettability of the liquid and external pressure.

A close inspection of the images shown in Figure 4b show that the time resolution was not sufficient to freeze the very fast motion of the moving contact lines at pore size level even at the fastest possible imaging speed using the monochromatic beam (here about 20 s per tomography in 16 bunch mode). Further optimizations and using the synchrotron in multi bunch mode we expect to reach an acquisition time for a tomogram of about 2 s, which will be sufficient to study slowly driven liquid fronts on the pore size level. To image the liquid fronts at realistic driving velocities as used in oil extraction we will need to improve the temporal resolution by another order of magnitude using a multilayer monochromatior.

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2. **Scientific Case & development aspects** (continued): provide details of the experimental programme with technical and scientific milestones**:**

Year 1

In the first year we will further develop the setups for the 3D stereo x-ray particle tracking technique. We will also develop the shear cell experiment and expect to capture the first data of trial runs. We will also develop the numerical schemes to analyze the data preferentially using MathLab codes. The insights gained during the data analysis will be feed back to optimize the experimental setups and the experimental parameters. Besides that we will start the preparations for the multilayer and hope to implement it beginning of year 2.

Year 2

We will implement the multilayer monochromator and expect that the setups for the stereo imaging technique and for the shear cell experiment are finished and might only require small optimizations and that we start collecting reliably data on the 3D motion of a fluidized granulate and the reorganization of packing geometry and liquid within sheared granulate. After careful analysis of the first data, fine tuning of the experimental parameter and of the experimental setups is expected to be needed. In case of the stereo imaging technique we will preferentially vary the agitation frequency and amplitude and keep the liquid content and the grain size constant. In case of the shear experiment we aim at various shear rates, shear amplitudes, and various liquid contents. Parallel to these tests we shall design and test a microfluidic setup to study driven liquid fronts in porous media. The porous media will consist of densely packed spherical beads. This matrix is chosen for its simplicity and the possibility to compare experimental with numerical results. Numerical schemes will be optimized and adapted to the experimental data to analyze the shape of a driven liquid front. By the end of the second year we expect first reliable results on liquid fronts with various driving speed.

Year 3

Within the third year we expect to complete our datasets on the fluidized granulates by varying the wetting liquid, diameter and mass density of the used tracer beads. This is expected to provide insight into the impact

on the liquid surface tension on the dynamic behavior, in particular the fluidization threshold and insight into the dynamic behavior of the Brazil nut effect for wet granulates. We expect to supplement the data sets of sheared granulate by varying the grains to aim at different wettability, surface roughness, and absolute pressure. This should provide insight into the different reorganization of liquid depending on the wetting angle and its potential impact on the altered reorganization of the packing geometry of sheared granulate. Variation in surface roughness and absolute pressure will provide insight in the importance of friction between the grains. Furthermore, we expect results on driven liquid fronts in porous media. The diameter of the beads will be varied as well as the viscosities of the reservoir fluid and the driven fluid in various porous matrices.

3. Scientific competence of the Group, Bibliography:

(a) **Competence:** provide a summary statement of the areas of expertise of the key participants.

Ralf Seemann is an internationally renowned scientist working on wetting and dewetting phenomena of thin liquid films on structured solid and elastic substrates, structure formation in thin films as well as rheological properties of thin films and their deviations from bulk behavior. For achievements he received a stipend of the Mitsubishi Chemical Center, Santa Barbara, in 2001 and the research award of the city of Ulm in 2003. In recent years, he started projects on ‘open microfluidics’ and on the mechanical properties of wet granular matter, where he adapted ultrafast x-ray tomography to image time resolved (re-) arrangements of liquid within a complex solid matrix. These experimental studies are completed by a microfluidics approach where he studies the behavior of emulsions in confining geometries. Since 2007 he is a full professor at the Saarland University and has a second affiliation with the Max-Planck institute in Göttingen, where he runs a research group working on wet granular matter and microfluidics.

Stephan Herminghaus has a long track record of basic research in the field of liquids, wetting, and soft matter systems. In 1995, he received a Heisenberg-Fellowship for research on nano-fluidics. He initiated and co-ordinated a Priority Research Program on "Wetting and Structure Formation at Interfaces", which was funded by the German Science Foundation and lasted from 1998 until 2005. It united about 30 leading research groups all over Germany in a joint endeavor to understand the fundamental mechanisms governing the behavior of thin liquid films at solid interfaces. His own projects in this program were concerned with micro- and nanoscale wetting as well as pattern formation in dewetting, wetting of biological surfaces, and wet granular matter. In 2003, he became a director at the MPI for Dynamics and Self-Organization and head of its Department for Dynamics of Complex Fluids, which consists of about 35 scientists conducting research on granular matter, complex fluids in micro-systems, biological physics, and general aspects of systems far from thermal equilibrium. He has published 135 peer-reviewed papers, which in total are now cited close to 500 times a year.

Martin Brinkmann has a long standing record on wetting related research involving chemically and/or topographically non-trivial geometries. He is very well acquainted with lattice gas simulations and the software tool ‘Surface Evolver’, which allows the numerical computation of equilibrium shapes of various types of liquid interfaces. During an extended post-doctoral stay at the Interdisciplinary Research Institute in Lille (France), he studied wetting of topographically structured substrates in view of applications, such as micro fabricated spray devices for mass spectroscopy. In early 2005 he became a principal investigator in the department of Dynamics of Complex Fluids at the MPI for Dynamics and Self-Organization in Göttingen. Current research topics are dynamical aspects of wetting and de-wetting on surface topographies and wetting in complex surface geometries, in particular in disordered assemblies of simple geometrical objects.

Eckart Meiburg is Professor and former Department Chair in the Mechanical Engineering Department at the University of California in Santa Barbara. Professor Meiburg's research interests lie in the general area of

fluid dynamics and transport phenomena. His group primarily employs the tools of computational fluid dynamics and linear stability analysis, in order to obtain insight into the physical mechanisms governing the evolution of a variety of multiphase flow fields. His accomplishments have been recognized by the Presidential Young Investigator Award of the National Science Foundation, Election as Fellow of the American Physical Society, a Humboldt Senior Scientist Award of the Alexander von Humboldt Foundation, and a Gledden Fellowship of the Institute of Advanced Studies at the University of Western Australia.

Marco Di Michiel is Beamline Operator Manager at the High Energy Scattering Beamline ID15. He has worked for more than ten years in synchrotron x-ray imaging and diffraction. He has developed the ultrafast tomography and imaging setup installed at ID15A.

(b) **Relevant literature**: list≤ 5 key papers, with titles, from proposers and/or from literature, relevant to the scientific and/or technical aspects.

R. Seemann, M. Brinkmann, E. J. Kramer, F. F. Lange, and R. Lipowsky, Wetting morphologies at microstructured surfaces, *Proc. Natl. Acad. Sci. USA* 102 (2005) 1848

M. Scheel, R. Seemann, M. Brinkmann, M. Di Michiel, A. Sheppard, B. Breidenbach, S. Herminghaus, Morphological clues to wet granular pile stability, *Nature Materials* 7 (2008) 189

D. Geromichalos, F. Mugele, S. Herminghaus, Nonlocal dynamics of spontaneous imbibition fronts, *Phys. Rev. Lett.* 89 (2002) 104503

D. Geromichalos, M. Kohonen, F. Mugele, S. Herminghaus, Mixing and condensation in a wet granular medium, *Phys. Rev. Lett.* 90 (2003) 168702

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4. **Technical details:** description of the proposed technical implementation of the LTP, including experimental set-up, sample environment, special support, etc.

<u>Dynamics of strongly fluidized granulates:</u>

The samples we will use in these experiments will consist mainly of fairly monodisperse glass spheres with diameters ranging from 100 to 600 μm with sufficient polydispersity in order to prevent crystallization. The tracer particles will have a large x-ray absorption contrast, such as BaTi or lead glass grains, allowing for a clear distinction between the tracers and surrounding media. Silicone oil will be used as the wetting liquid since its' boiling point is above 250 ^{o}C and radiation damage was not observed in previous experiments. The grains will be prepared in small glass vials with a diameter of 8 mm and mounted on a vertical shaker, which can be installed on the rotation stage of the tomography setup. For agitation frequencies in the range of 200-1000 Hz the minimum amplitude required for fluidisation is below the diameter of a single grain so the entire granulate can be visualised over the course of the agitation.

Due to the improved flux generated by the new undulator source at ID15 (about 20 times at 50 keV compared to the wiggler) and using an efficient scintillator like LuAG:Ce, we reach exposure times down to 2 ms even using monochromatic x-ray beam. This will allow to split the beam into two and to capture images from two different view angles at the same time and to determine the 3D, x-y-z position of tracer beads at 2 ms time resolution. Using a multilayer monochromator a further decrease in exposure time by at least ten times is expected.

The undulator radiation is monochromatized by two bent Laue (111) Si crystals monochromators with asymmetric cut χ and the thickness T optimized to achieve the maximum integrated intensity in the monochromatic x-ray beam. The beam is then split in two beams of equal intensity using a third bent Laue monochromator which reflects part of the incident photons at an angle 2θ, (θ being the Bragg angle) and transmits the other (Figure 5a). The sample is positioned in the region where the two beams overlap and imaged using a single detector (Figure 5b). The detector will be positioned at a distance such that the two

beams are not overlapping. The detector records simultaneously two “half images” of the sample taken from two directions separated by the angle 2θ. Comparing the two “half images”, the xyz coordinates of the beads can be determined. Figure 5b shows a first preliminary result where two images were obtained from a split beam from a static granular sample using the Sarnoff camera.

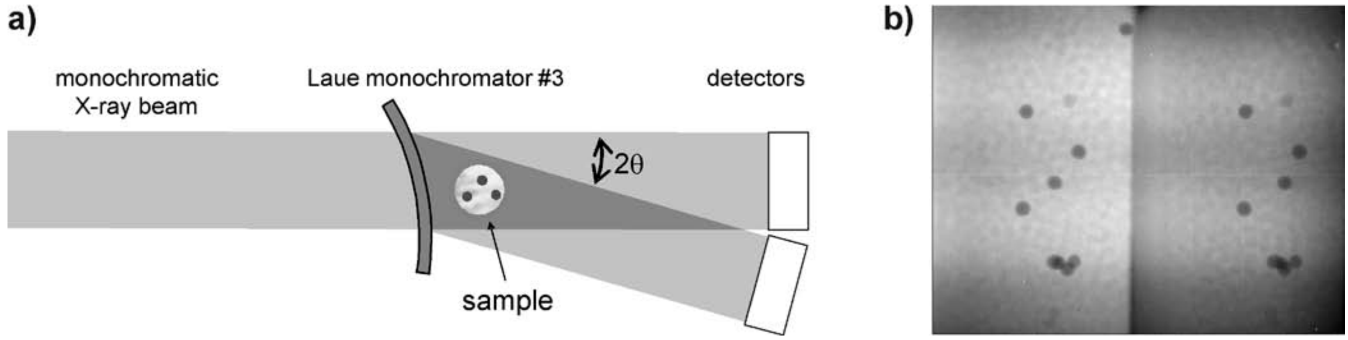

Figure 5 a) Sketch of the proposed stereo imaging technique. b) Proof of principle using the stereo imaging technique. Granular sample containing BaTi tracer particles is imaged from two view angles projected on the chip of a single CCD camera.

<u>Sheared granulate:</u>
The granular material consists of fairly monodisperse glass beads with a diameter ranging from 150 to 500 µm. The spread in bead size is from a few percent to about 20%, enough to prevent crystallization. The beads will be prepared in a small home build shear cell with a diameter of about 8.5 mm to fit to the beam width. The setup can be mounted on the rotation stage of the tomography setup. The shear force will be applied and measured by computer controlled microfluidic pumps and pressure sensors connected to the shear cell via hydraulic lines. The shear rates are in the range of 1^{-4} s^{-1} to 1^{-2} s^{-1} while the corresponding shear velocities range from 0.1 – 60 µm/s. Assuming a realistic time to capture a full 3D tomography to about 10 s for our system with the current setup (and down to about 1 s with the new CMOS camera), the time resolution should be sufficient to freeze the motion of the beads, the stretching and reformation of capillary bridges and liquid clusters for the smallest and moderate shear velocities. Thus, the dynamic rearrangements within a sheared wet granulate can be studied with good time resolution using fast x-ray tomography. Using the multilayer monochromator we expect to image at shear velocities above 20 µm/s. As the wetting liquid we will use water with a significant amount of dissolved NaI or ZnI_2 to increase the x-ray absorption contrast. The different absorption allows for a clear distinction between the air, the glass beads, and the liquid.

<u>Driven liquid fronts in porous media:</u>
As porous matrix we will mainly use cylindrical vials filled with spherical beads ranging from 50 to 500 µm with a fairly homogeneous size distribution. Compared to natural porous media like sandstone the x-ray contrast is much more homogeneous and enables a clear separation of the liquid, the solid matrix and the air and therefore a precise analysis of the 3-D-tomograms. Furthermore, the results are expected to be compared to results from numerical simulations and to the equilibrium distribution of a certain amount of liquid in a reservoir. To explore driven liquid fronts, the bottom of the sample vial will be connected to a syringe via teflon tubing whereas the top will be connected to a pressureless reservoir. A syringe pump will applying a constant flow to the sample. Experiments with various pore (bead) sizes and liquid viscosities will be performed to analyze the liquid front and complex internal structure with good statistics. Beads with larger size will be used to study the exact shape of the liquid front as well as to study the emergence and the exact shape of liquid inclusions, their dynamic behavior when the liquid front has passed. As a reservoir liquid we will use oil (typically silicon oil since the viscosity can be easily varied without changing the wettability). As a driving liquid, we will use water with a significant amount of solved NaI or ZnI_2 to adjust both, the x-ray absorption and the viscosity. The different absorption allows for a clear distinction between air, the glass beads and the liquid. To increase the complexity of the porous matrix but to keep the homogeneous x-ray absorption, we might also use sintered glass filters or special sand stones.
We expect to image liquid fronts with front velocity of several micrometer per second on pore sizes of about 5 µm using a multilayer monochromator, corresponding to typical driving speeds in field oil extraction.

5. **Beamtime Request:** please begin with preferred starting period

Scheduling period	**Beamline(s) Requested**	**Shifts requested**	**Results expected (cf. Milestones, §2)**
2009/II	ID15A	18	Installing the setup for the stereo imaging technique. First tests of the shear cell and the setup for the particle tracking of a strongly fluidized granulate.
2010/I	ID15A	18	Optimizing the experimental setups of the stereo imaging particle tracking technique and the continuous recoding of fast tomograms with the shear cell experiment. Starting the preparations for the multilayer monochromator.
2010/II	ID15A	18	Installing and testing the setup for the multilayer monochromator. Recording of flight paths of strongly fluidized granulates varying frequency using the multilayer monochromator. Recording data sets of sheared granulate for various shear rates and shear amplitudes. Tests of the microfluidic setup to study driven liquid fronts.
2011/I	ID15A	18	Fine tuning of the experimental setups. Recording flight paths of strongly fluidized granulates varying the amplitude. Recording data sets of sheared granulate for various liquid contents. Measuring driven liquid fronts for various driving speeds.
2011/II	ID15A	18	Completing data sets for fluidized granulates by varying the wetting liquid and bead diameter. Completing data sets for sheared granulate for various wettability. Measuring driven liquid fronts for various viscosities.
2012/I	ID15A	18	Completing data sets for fluidized granulates by varying the wetting liquid, bead diameter and mass density of the used tracer beads. Completing data sets for sheared granulate for various surface roughness of the tracer beads and absolute pressure within the shear cell. Measuring driven liquid fronts for various pore space geometries.
			We request for 18 shifts per beam time since the proposed experiments are quite time consuming regarding the experimental setup and the experiment itself: Each run requires about 4-5 hours for sample alignment and data acquisition. Even that we use high speed imaging to freeze the motion of e.g. of a liquid front we will continuously capture tomograms for about 2 hours per data set.

6. Resources

(a) What resources and services are expected from the ESRF ?

Sarnoff and Dalsa camera and CMOS camera. 1x and 5x optics

(b) What resources, not available today, are required to be provided or developed by the ESRF (staff, instrumentation) ?

Stereo x-ray particle imaging velocimetry, will be done in collaboration with proposer.
CMOS camera with 1k x 1k pixel, 1000 fr/sec, which will be purchased by ID15 during the year 2009
2 x optics would be welcome but is not mandatory.

(c) What contribution is the User group bringing to the ESRF in support of the LTP (financial, human, technical resources) ?

We plan to hire up to 10 postdoctoral researchers/PhD students to support the experiments. One postdoctoral researcher should be affiliated at the ESRF to support the method development.

We are willing to pay for the costs of a multilayer monochromator and /or new optics up to 50 k€ and substantially support its implementation.

We will further develop stereo imaging and its numerical analysis, which could be provided to other users.

(d) Indicate how and when the resources listed in 6(c) will be obtained (in-house resources, grants…)

Postdoctoral researcher (Alexander v. Humboldt Foundation, running): Zeina Khan
PhD student (German Science Foundation, graduate school 1276, running): Somnath Karmakar
PhD student (core funding Saarland University, running): Mario Scheel
Two Diploma students located at the Saarland University, running: Marc Schaber, Michael Jung
Postdoctoral researcher located at the ESRF: funding requested from BP (decision expected in March): n.n.
2 Postdocs/PhD students located at the Saarland University: funding requested from BP (decision expected in March): n.n.
Postdoctoral researcher (funding requested from A. v. H foundation otherwise core funding Saarland University) (running from May): Jean-Baptiste Fleury
3 Postdoctoral researcher/PhD students located at the MPI-DS (funding requested from BP otherwise MPI core funding) (positions announced)

The cost for the multilayer monochromator and/or optics will be split between the MPI for Dynamics and Self-Organization and the Saarland University.

The stereo imaging and its numerical analysis will be further developed from Marco DiMichiel, Zeina Khan and one n.n. postdoctoral researcher/PhD student.

7. **Expected long-term benefits to the ESRF User Community** such as new instrumentation, new techniques, new areas of research with potential user base.

More than ten time increased x-ray flux in the monochromatic beam at around 50 keV thanks to the installation of a high energy multilayer. This is expected to be useful for most of the tomography-imaging users. In fact several experiments, performed at the moment using white x-ray radiation for reasons of flux, could be performed in monochromatic beam. So allowing a better optimization of the signal-to-noise ratio and a substantial reduction in sample radiation damage

Fast dynamics of organic phases in porous media, in particular the technique to continuously image liquid fronts in porous media is expected to find a large potential interest in new user groups:

3D x-ray particle tracking with sub millisecond time resolution. Particle tracking allow to answer questions regarding any particle dynamics and thus e.g. to mixing behavior. But moreover, it might also allow to measure flow velocities of liquid in confined geometries seeded with tracer particles by tracking the motion of the seed particles. This might allow also visualizing flow patterns in e.g. plants. The numerical code to trace the 3D position of the particles will be provided to other users.

Time resolved imaging of sheared granulate should be also suitable to explore the mechanical behavior of slurries and suspensions. Given the fact that about half of the raw materials come as a solid and are thus processed as granulate these questions are not just of fundamental scientific interest but also have a large impact on industrial processing. Among them, mixing, mechanical stability, and material transport are probably the most important ones. This applies to e.g. to concrete, food industry, pharmaceutical industry, or geology.

Being able to image liquid fronts with good time resolution should be interesting to answer to a huge range of different fundamental questions like studying effects describe by Washburn law or Darcy’s law on a pore size level and not on the level of an average porosity. Applications of these questions can be found e.g. in impregnation, liquid infiltration: causing e.g. frost damage of streets and houses, storage of nuclear reactive waste, agriculture, environmental geology (cleaning of soil from hydrocarbons), water reservoirs, oil mining, liquid transport in fuel cells, engineering of filter cakes, water separation (gas/chemical processing), water evaporation from reservoirs, spreading of liquids into (woven and nonwoven) fabrics: printing techniques, polymer injection molding, fiber-reinforced polymers,.......

Annex to long term proposal

"Dynamic distribution of liquid in fluidized granular matter and porous media:
from mixing to oil extraction"

The ESRF long term proposal "Dynamic distribution of liquid in fluidized granular matter and porous media: from mixing to oil extraction", is embedded into a research grant from British Petroleum (BP) to develop a detailed scientific understanding of the basic physical and chemo-physical properties with the aim to develop schemes for enhanced oil recovery. This research project was meanwhile approved by BP and will start presumably from June 2009.

In that framework, BP funds three research groups within Europe with a financial volume of together 3 million USD per annum, working on granulates and driven liquid fronts. The projected running time of the project is ten years. With an renewal process after the first five years.

The research group located in Göttingen/Saarbrücken with the principal investigators R. Seemann, S. Herminghaus, M. Brinkmann, and E. Meiburg, who also applied for the ESRF long term proposal together with M. Di Michiel (ESRF), is working on the structural properties of granulates (sediments and consolidated rocks) and the static distribution of liquid within granular systems and driven liquid fronts both experimentally and theoretically. A significant part of the experimental research program will be done using the ESRF ultrafast x-ray microtomography setup at ID15A.

All the claimed positions for postdoctoral researcher and PhD students mentioned in the ESRF long term proposal were approved. In particular the postdoctoral researcher located at the ESRF can be hired presumably from June 2009. Thus, all the financial and human resources are guaranteed to make the ESRF long term proposal a real success.