

Colloidal dynamics in a glass forming solvent at low temperatures

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We have measured the dynamics of colloidal nano-particles in a glass forming solvent by means of X-ray photon correlation spectroscopy (XPCS). In order to follow the dynamics close to the temperature induced glass transition, we have used a mini cryostat allowing temperatures down to 80K with a very high stability (~ 0.01 K).

Our measurements have been performed on a dilute suspension of silica particles ($D=0.5$ micron) in propanediol, a liquid that goes through a glass transition at $T_g = 170$ K. By using the Medipix detector, a 2-D pixelated detector with a full frame rate up to 30 Hz, it has been possible to quantify the dynamics in the temperature range 240K-205K.

Interestingly, though the solvent is still far from the glassy state, our data clearly show that the colloidal dynamics change around $T = 215$ K (figure 1 and 2). At “high” temperatures the particle dynamics is Brownian, with a dispersion relation Γ proportional to Q^2 and simple exponential decay of the correlation functions. By decreasing the temperature, we obtain compressed exponential correlation functions ($\gamma > 1$) and simultaneously, a gradual change occurs in the power-law of the dispersion with an exponent approaching ~ 1 at $T=205$ K.

Measurements on suspensions with higher volume fraction suggest that the dynamics below ~ 215 K is collective and our results can be explained by a transition from continuous to intermittent dynamics, probably driven by stress relaxations of the solvent [1].

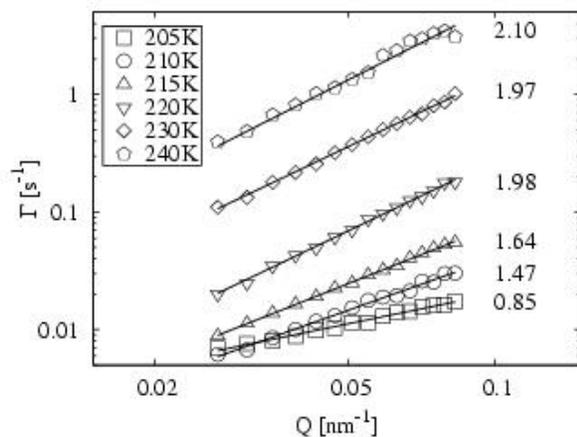


Figure 1 Dispersion relation at various temperatures. Lines are guide to the eye

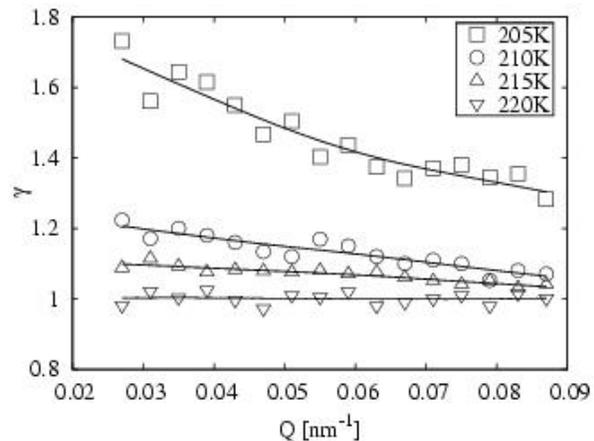


Figure 2 Compressing exponent vs Q at various temperatures. Lines are guide to the eye

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The SrTiO₃ displacive transition revisited by Coherent X-ray Diffraction

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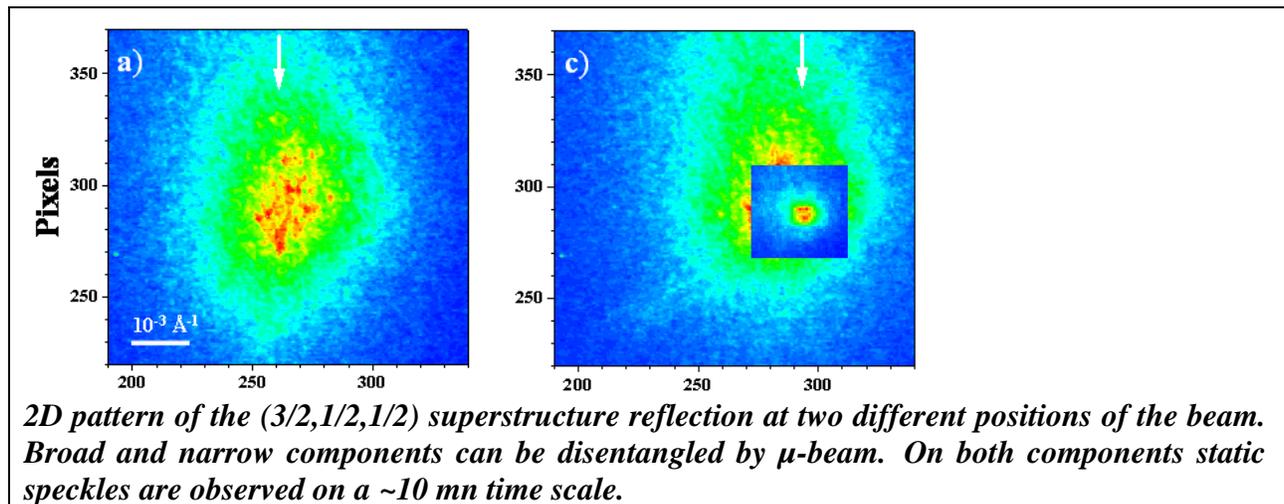
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Although most issues concerning the application of the scaling theory of phase transitions have been settled long ago, two frequently observed scattering features remain unaccounted for within standard scaling theory: the "neutron" central peak (CP) and the "x-ray" narrow component (NC). Remarkably, both features were first evidenced in studies of the critical behavior associated with the $T_c \sim 100\text{-}105$ K antiferrodistortive transition in the perovskite SrTiO₃. We present a Coherent X-ray Diffraction study of the antiferrodistortive displacive transition of SrTiO₃. From the microbeam x-ray coherent diffraction patterns, we show that the broad (short-length scale) and the narrow (long-length scale) components can be spatially disentangled. Moreover, both components exhibit a static speckle pattern. This gives evidence that the narrow component corresponds to static ordered domains. We interpret the speckles in the broad component as due to a very slow dynamical process, corresponding to the well-known central peak seen in inelastic neutron scattering. Beyond this experiment, we show that CXD is a valuable new tool to study phase transitions and defects in the low temperature ordering.

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XPCS studies of slow, non-diffusive dynamics in glassy soft matter

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For many disordered soft solids -- such as foams, gels, concentrated emulsions, and dense colloidal suspensions -- the solid phase is separated from a fluid state by an ergodic-nonergodic transition that leaves the system in an out-of-equilibrium configuration. Often such out-of-equilibrium materials display a protracted evolution of their dynamical properties that bears strong resemblance to the phenomenon of aging seen in molecular glasses and glassy polymers. Experiments on aging have typically measured the temporal evolution of response functions, such as the elastic moduli, and far fewer studies have characterized dynamical correlation functions in aging systems. In an effort to understand the microscopic dynamical behavior of out-of-equilibrium disordered materials, we have conducted x-ray photon correlation spectroscopy (XPCS) studies on a series of disordered soft solids. The combination of large wave vectors and long time scales accessible with XPCS makes the technique uniquely well suited for elucidating the nanoscale motions in such glassy materials.

The systems we have investigated include suspensions of the synthetic clay laponite [1], which slowly evolve from fluid to solid in a manner akin to aging, as well as concentrated nanoemulsions [2] and depletion gels [3], whose dynamical evolution we tracked after the cessation of a strong, fluidizing shear. In the studies involving shear, our motivation was to compare the dynamical recovery of a jammed system following shear flow with aging in glasses, which jam through a quench in temperature. For all the disordered soft solids, we observed the same generic slow dynamics characterized by an intermediate scattering function that follows a “compressed” exponential form, $g_1(q,t) \sim \exp[-(t/\tau)^\beta]$, with $\beta \approx 1.5$ and $\tau \sim q^{-1}$. Such behavior contrasts with glassy diffusion, for which correlation functions are stretched ($\beta < 1$) and $\tau \sim q^{-2}$. Thus, we conclude that the dynamical evolution of the disordered soft solids, while displaying signatures of aging, cannot be directly related to traditional aging phenomena in glasses.

Instead, these results indicate a type of slow, non-diffusive dynamics that are apparently universal to a range of disordered soft materials. In particular, our observations can be understood in the context of a recent model that describes the dynamics in disordered elastic media in terms of strain from random, highly localized stress relaxation events. Within this picture, the constituents of the jammed systems move with a broad distribution of strain velocities. This talk will provide an overview of these studies with an emphasis on recent work to uncover the microscopic origins of this non-diffusive motion. In particular, insights gleaned from further examination of the XPCS results, including analysis of the instantaneous two-time correlation functions for these evolving systems, will be discussed.

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Diffusion Studies with X-Ray Photon Correlation Spectroscopy at Present and Future Light Sources

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The dynamical properties of hard and soft condensed matter are of considerable importance from a fundamental point of view, but are in addition essential for the technical applications of bulk or nanoscale materials. Macroscopically, diffusion can be mathematically described (Fick's laws) and investigated with standard methods like tracer diffusion experiments, for a review, see Mehrer [1]. However, the underlying processes on an atomic level, i.e. atomic jump vectors and frequencies, are not easily accessible. Up to now only quasi-elastic methods, particularly quasi-elastic neutron scattering (QNS), quasi-elastic Mößbauer spectroscopy (QMS), nuclear resonant scattering (NRS) of synchrotron radiation, and (most recently) the neutron-spin echo technique, were capable of resolving the elementary atomic jump process (for a review, see Vogl and Sepiol [1]). The above mentioned techniques suffer, however, from two big disadvantages. First, only certain isotopes are “visible” for each technique, e.g. the Mößbauer isotope ⁵⁷Fe in case of QMS and NRS. Second, due to a limited energy resolution of all techniques only very fast diffusion is detectable by the above-mentioned methods. Hence, a non-resonant technique – not restricted to certain isotopes–, which can detect slow diffusion, is extremely desirable. The emerging method of X-ray photon correlation spectroscopy (XPCS) [2] is the most promising candidate to fill this gap. Basically the accessible wavelength and coherency of modern 3rd generation synchrotron sources should be sufficient to resolve the jump process on the atomic level, however, as the coherent intensity in the hard X-ray regime is limited, one has not succeeded until now in experimental verification. Having the forthcoming extremely brilliant fourth-generation sources at the horizon, a serious assessment of possible diffusion studies seems to be mandatory. This next generation of sources, based on a free-electron laser, is already operating and will culminate with the hard X-ray FEL's expected to become operational near the end of this decade. Here we present a theoretical treatment of the expected signal-to-noise ratio given a certain count rate, that is we deliver estimates on the minimum count rates for measuring atomic diffusion and present recipes for optimising the signal-to-noise ratio. Also we examine samples concerning their expected scattered intensity and the possibility of resolving atomic diffusion at today's synchrotrons. Furthermore, an outlook is given on the possibility of such measurements at the European X-ray free electron laser [3].

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Synthetic incoherence: Gaussian confinement of the mutual coherence function (Part A) and Dynamical diffraction: diffraction with energy loss in the plasmon regime of crystalline silicon (Part B)

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Part A. An incoherent signal is often desirable for imaging and tomography, but available sources in electron and synchrotron x-ray microscopy are often highly coherent. Here, I discuss theoretically a method to achieve a mutual coherence function with Gaussian confinement. The length scale of the region significantly different from zero is in the nanometer range for a typical application in transmission electron microscopy. Two schemes are presented: in one, a flood beam is scanned over the sample using a 2D rectilinear Gaussian pattern; in the other, a flood beam is scanned over the sample using “tapered-cone illumination”, a term introduced here. Tapered-cone illumination is a 2D generalization of hollow-cone illumination in which the cone angle is varied and weighted with a Gaussian. Analytic and numerical results are given in all cases; the mutual coherence function in tapered cone illumination is seen to be much more confined than hollow-cone illumination, which has a $1/\sqrt{R}$ tail, where R is the separation of the two points.

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Part B. Dynamical diffraction refers to the process of simultaneous energy loss and diffraction in transmission electron microscopy. In the present work, the classic formulation of Kohl and Rose is implemented numerically. A wave functions of a 50 keV electron entering and exiting a silicon crystal via the (111) face are found. The crystal potential is represented as a linear combination of Hartree-Fock atomic potentials. The incoming and outgoing electrons interact via the mixed dynamical form factor of the crystal, which, in turn, is given in terms of the nonlocal, energy-dependent dielectric function. The dielectric function is computed within the local-density functional formalism using the Random Phase Approximation. Figures are presented representing the diffraction patterns with energy losses of 10 eV to 25 eV in 2.5 eV steps, i.e., across the plasmon regime. If the incident beam is in the [111] direction, i.e., normal incidence, six-fold diffraction spots are predicted to be reasonably intense right at the plasmon energy, but much less intense away from the plasmon energy. For oblique incidence directions, lower symmetry diffraction patterns are predicted, with diffraction spots arising in a minority of cases.

Focusing Applied to Small-Angle Multispeckle XPCS Experiments

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XPCS performed with area detectors, so called multispeckle XPCS, has recently been extended to delay times as short as a several milliseconds [1,2] using fast and efficient direction-detection area detectors. But increasingly short delay times and the coherent flux limitations of third generation synchrotron sources require optimal use of the delivered undulator brilliance. For large collimating slit sizes, it has been shown [3] that the signal-to-noise ratio (SNR) for a measured autocorrelation decay function is maximized when the angular extent of the detector pixels as seen from the sample position matches that of the source as seen from the sample position. At a third-generation storage-ring x-ray source like the APS with a highly asymmetric source, this criterion presents an immediate problem because area-detector pixels are typically square. It is natural, therefore, to employ focusing optics to tailor the effective source sizes so that the resulting x-ray speckles approximately match the size of the detector elements. But since it is easy to degrade the effective brilliance of the beam via imperfect optics [4] it is also important to carefully characterize the effect of such optics on the coherence and stability of the x-ray beam. Here we report characterization of small angle x-ray speckle produced by a pinhole set-up, a Fresnel zone plate (FZP) set-up (and, possibly, a one-dimensional kinoform lens set-up). As compared to the pinhole set-up, we find for the FZP set-up that the speckle amplitude is approximately preserved but that the speckle widths are considerably greater and the signal per speckle is greatly increased. We compare our measurements to calculations. Our results demonstrate that strongly demagnifying optics like FZP's and kinoform lenses can be employed to produced virtual sources that fully utilize the brilliance delivered by third generation x-ray sources.

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Towards Soft X-ray Photon Correlation Spectroscopy of Critical Fluctuations

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The use of coherent soft X-rays for resonant magnetic scattering provides numerous possibilities for the study of *magnetic* nanostructures. For instance, static and slowly varying magnetic domains can be imaged, in particular, by holography in combination with resonant scattering [1]. In this way the reversal behavior in applied magnetic fields of extended and nanostructured magnetic objects has been investigated [2,3].

However, measuring the *dynamics* of magnetic samples remains an elusive and challenging task. Combining soft X-rays with the method of photon correlation spectroscopy (sXPCS) opens the opportunity of measuring such dynamics. While the photon correlation technique is routinely employed using visible light or hard X-rays, only few pioneering experiments have been carried out with soft-X-rays [4], and so far never in order to study magnetic fluctuations.

In the case of magnetic systems, the fluctuating units are regions with locally correlated magnetization, so-called spin blocks. By exploiting the x-ray magnetic dichroism as a unique contrast mechanism, sXPCS studies promise to be the first direct probe of spin blocks, without having to resort to macroscopic properties, such as magnetic susceptibility. The measurement of scaling law exponents close to magnetic phase transitions will be of fundamental interest.

Here we present first results of sXPCS experiments towards this goal that were performed at the BESSY II synchrotron source in Berlin, Germany. For pilot studies, we produced single perpendicular anisotropy layers with a Curie temperature slightly above room temperature and sample systems with size controlled magnetic entities to investigate critical magnetic fluctuations and the transition to a superparamagnetic state. Here, we report on recent progress of this project.

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Coarsening Dynamics of Au Precipitates in a Thin Fe Film

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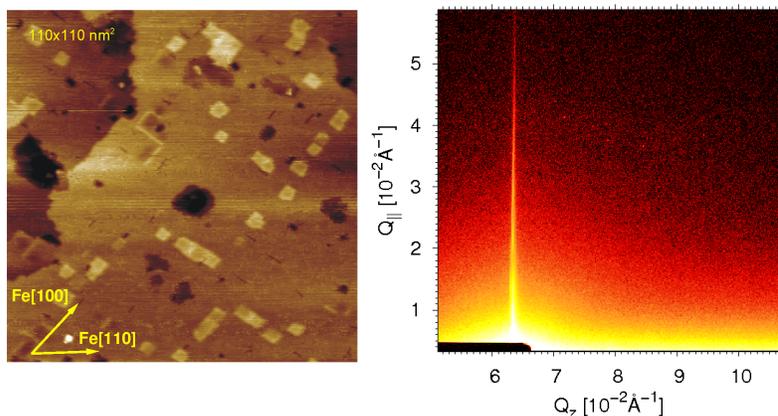
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Iron films grown on a Au(100) substrate are one of the best known examples for self-surfacing action promoting a flat 2D-growth [1]. During iron evaporation gold atoms are segregating onto the iron surface. As a result the iron film is immediately covered by a floating Au layer whose surfactant effect induces the layer-by-layer growth mode of the iron film. At elevated temperatures the segregation process is enhanced and Au precipitates are formed within the iron layer. Due to the strongly differing atom sizes between Au and Fe, lattice strains are introduced, causing plate-like precipitates aligned along Fe<100> directions.

The precipitate growth and coarsening can be monitored *in situ* by changes in the grazing-incidence small-angle X-ray scattering (GISAXS) diffraction pattern. Furthermore, by combining GISAXS with the method of X-ray photon correlation spectroscopy (XPCS) it becomes possible to study the slow dynamics of the coarsening process near the surface.

In order to analyze the fluctuating speckle intensities the detrended fluctuation analysis (DFA) was applied [2,3]. The DFA was performed for different scattering vectors (Q_{\parallel}) with the incident X-ray beam parallel to the Fe[100] and Fe[110] direction, respectively. Experimental results indicate stronger coarsening dynamics along Fe[100] directions. This finding suggests that coalescence of neighboring precipitates plays a leading role for the present coarsening mechanism



Left: STM topograph of a 10ML Fe film on Au(100) after 1h of annealing at 610°C. [1] **Right:** GISAXS diffraction pattern with pronounced speckles of an identical sample annealed for 4.5h at 650°C.

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Cluster based Analysis for Multi-Speckle XPCS Experiments

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Using fast and efficient area detectors has extended the range of accessible timescales in multi-speckle X-ray Photon Correlation Spectroscopy (XPCS) experiments to the millisecond regime [1,2] and with the plans to build even brighter x-ray sources it seems feasible to investigate dynamics of microsecond timescales with multi-speckle XPCS in the near future. However, data rates are approaching GB/s levels and the ratio of data collection time to data reduction time has become very unfavorable, e.g. a standard data set of 1000 frames might be collected in ~10s or less and its analysis would take ~1000s or more.

The data reduction time increases linearly with the number of pixels involved. This suggests that the multi-tau algorithm to calculate the intensity-intensity auto-correlation function g_2 is highly parallelizable. We have implemented a set of C⁺⁺ routines to perform the multi-speckle XPCS analysis on a computer cluster. In a first step, we interfaced the cluster based g_2 calculation with the existing Matlab based code to analyze XPCS data in a post data collection mode. First tests show strongly improved data analyzing times. In addition, we will present a roadmap towards (near) 'real time' multi-speckle g_2 calculations.

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Stationarity and Spatiotemporal Dynamics at the Orbital Ordering Transition in the Half-doped Manganite $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$

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Understanding the mesoscale structure and dynamics of complex charge, spin, and orbital-ordered phases is crucial to unraveling the mystery of the colossal magnetoresistance (CMR) phenomenon. The use of coherent soft x-rays allows this complexity to be mapped into far-field speckle-diffraction patterns, which can be analyzed to probe spatiotemporal dynamics, microscopic memory effects, and real-space structures through phase retrieval. We report new coherent soft x-ray scattering experiments performed on beamline 12.0.2.2 at the Advanced Light Source wherein we probe the structure and dynamics of orbital domains in $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ near the orbital ordering transition temperature. By operating at the Bragg condition for the lowest-order orbital reflection and at a wavelength that is resonant with the Mn L-edge, we have obtained excellent scattering signal with direct sensitivity to the orbital ordered phase. We find that the low temperature state is characterized by static short-range ordered domains and that the system remains largely static as it is warmed through the disordering transition. In addition, we have observed small-amplitude spatiotemporal fluctuations near the transition. We have studied the connection between this small fluctuating component, a decrease in the orbital domain coherence length, and a change in shape of the integrated scattering profile over a narrow temperature range all just below the transition. Our measurements present new insight into how the orbital ordering domain state behaves near the order-disorder transformation - a clue into a key ingredient of the CMR story.

Hydrodynamics of concentrated fluid hard-sphere colloids probed by X-ray Photon Correlation Spectroscopy

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We present an extensive X-ray scattering study of the static and dynamical properties of hard-sphere suspensions of PMMA particles at increasing volume concentrations close to the freezing limit ($\phi=49\%$). The static structure factors have been extracted via Small-Angle X-ray Scattering measurements while the dynamic behavior has been probed by X-ray Photon Correlation Spectroscopy (XPCS), the X-ray analog of Dynamic Light Scattering (DLS) that quantifies temporal correlations in dynamic systems by looking at the fluctuations of the scattered intensity under coherent illumination. The hydrodynamic functions resulting from the short-times diffusion coefficients are discussed in the frame of the δ - γ expansion model of Beenakker and Mazur[1] by using the ideal hard-sphere potential and by using the measured structure factor. In addition, the functional forms for the dynamic structure factors are discussed and compared to previous DLS work[2,3] and another XPCS study that does not agree about a common scaling behavior at the long times when approaching the freezing concentration [4].

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Coherent X-ray Resonant Magnetic Scattering studies

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Coherent X-ray Resonant Magnetic Scattering (C-XRMS) is a unique tool allowing the investigation of nanoscale structural and magnetic domain topologies in complex materials. While the standard scattering technique gives information about averaged long range order the use of a spatially-filtered coherent X-ray beam implies interference effects between the scattering paths and therefore offers in addition information about specific local disorder, in other terms the specific morphologies¹. Furthermore the use of resonance effect by tuning the energy of the light to absorption edge of studied element gives a specific sensitivity to magnetic structure in the material². The wavelength of the light, in the soft X-range is perfectly suitable to study structural feature at the nanoscale. The resulting coherent scattering pattern, detected on a bidimensional detector, exhibits speckle feature, which can be defined as a *magnetic speckle pattern* as it gives an indirect image of the specific magnetic topologies and can be used for correlation studies³.

We present here results obtained on perpendicular exchange bias thin films made by stacking ferromagnetic Co/Pt multilayers with antiferromagnetic IrMn layers⁴. These systems exhibit a perpendicular magnetization arising from the ferromagnet and leading to striped domain structures in the plane of the film. Exchange coupling effects appear at the interface between the ferro and antiferromagnetic layers while cooling the system down, below a blocking temperature. Our C-XRMS studies on these systems in transmission geometry give a unique insight on the influence of the exchange coupling on the behavior of the magnetic domain morphology in the ferromagnet under magnetization process. By acquiring speckle patterns along the hysteresis magnetization loop, and correlating the patterns together, we could evidence that the ferromagnet acquires a strong magnetic domain memory through exchange coupling effects with the antiferromagnetic layer. This phenomenon is interpreted by the formation of a reference magnetic template in the antiferromagnetic underlayer during the cooling process. Most importantly, we have devised a procedure by which this domain memory can be tuned by varying the sample cooling protocol.

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Dynamics during a transient gelation process studied by XPCS

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Aggregation and gelation are topics of fundamental interest in condensed matter physics that also have many direct industrial applications. Transient gelation is a non-equilibrium phenomenon (also known as *delayed sedimentation*) that can be found in colloidal suspensions with strong-enough short-ranged attractive interactions. Due to the interactions, the colloidal particles can aggregate at much lower concentrations (20 %) than those leading to the formation of glasses in hard-sphere systems (60 %), to form a space-filling structure often denoted a gel. However, this non-equilibrium structure slowly evolves, until the spatial connectivity is lost and the gel suddenly collapses.

A common generic behavior of disordered soft-matter systems such as colloidal glasses and gels is the presence of several dynamical relaxation mechanisms. The fast(er) ones correspond to the confined motion of individual particles or aggregates in cages or clusters created by neighboring particles/aggregates. Because the clustering can lead to structural arrest, such systems are generally non-ergodic. It is through the slow relaxations, corresponding to structural rearrangements equivalent to the α process in glasses that the system eventually can reach equilibrium and ergodicity may be restored.

In this paper we study the slow, non-equilibrium dynamics during transient gelation by X-ray photon correlation spectroscopy (XPCS). The intermediate scattering functions change during the process from stretched to compressed exponential decays indicating a jamming of the system in the full aging regime. A complex aging behavior towards the final collapse of the gel is observed and we propose that large scale network deformations trigger an un-jamming process leading to the collapse.

X-ray Photon Correlation Spectroscopy in Microfluidic System

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We present a new experimental method that combines X-ray photon Correlation Spectroscopy (XPCS) and microfluidics and allows the direct measurement of the mesoscale dynamics of various soft matter systems (e.g. colloids, polymers, biological molecules like proteins, RNA, etc.) under flow. XPCS is an ideal way to perform direct measurements on the underlying slow (10^{-3} - 10^3 s) mesoscale (~ 100 - 5000\AA) dynamics in a large class of hard- and soft- condensed matter systems. Microfabricated fluid mixers are being used nowadays to study micro-scale flow regimes both with the purpose to achieve new functionality or as an experimental tool. Our aim is to develop an experimental method that combines microfluidics and XPCS in order to measure the intrinsic mesoscale dynamics taking place in the fluid. Such a setup reduces the risk of beam damage and also allows time-resolved studies of various processes taking place in mixing flowcells. In the experiments reported here, we have used colloidal suspensions of hard-sphere systems and studied their Brownian dynamics while flown through in-house made flowcells.

Our experimental results agree with theoretical predictions, and show that in the low-shear limit and for a transverse scattering geometry (scattering vector q perpendicular to the flow) the diffusive dynamics of the PMMA particles is decoupled from the flow-induced convective response. In such a case, the homodyne intensity fluctuation correlation times measured by XPCS scale with q^{-2} as expected for a suspension of non-interacting Brownian particles, and are independent of the flow rate. However, this result does not hold for more general geometries. The correlation times measured in a homodyne-XPCS experiment depend on the Brownian diffusion time, on the shear rate and also on the exact scattering geometry. Here we show theoretical predictions and experimental results for XPCS measurements performed in a longitudinal geometry (scattering vector q perpendicular to the flow).

Critical magnetic fluctuations measured in the time domain with coherent x-rays

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In recent experiments the magnetic structures of thin holmium films of various thicknesses have been characterized with x-ray resonant magnetic scattering at the holmium M₄₅ resonance. Over a 15-20 K wide temperature interval around the magnetic phase transition, the magnetic superstructure peak was found to display critical broadening, with signs of a crossover from three to two-dimensional behavior. In experiments performed at the Berlin synchrotron BESSY we directly measured the magnetic fluctuations that arise at this phase transition by applying x-ray photon correlation spectroscopy (XPCS). By filming the speckle pattern with a CCD detector, we can directly observe this magnetic phase transition, as a change from static at low temperature to dynamic speckle at higher temperature. The observation of a static speckle pattern on top of the dynamic speckle shows that the system is non-ergodic and pinned to the defect structure of the film. With this study we proof the feasibility of resonant XPCS to address the low-frequency dynamics of sub-micron magnetic correlations.

Surface Dynamics investigated with X-ray Photon Correlation Spectroscopy

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X-ray photon correlation spectroscopy used at grazing incidence angles is a powerful tool to study slow surface dynamics of soft matter systems. It is highly surface sensitive, element specific and it overcomes the limitations of dynamic light scattering regarding multiple scattering and accessibility of large length scales only. XPCS has been used successfully to investigate capillary wave dynamics in various aspects, like e.g. on bulk liquids, at the freezing in of surface dynamics close to the glass transition, or dynamics of polymer films in confined geometries. Recently, we demonstrated that slow surface dynamics on nano meter length scales can be investigated with XPCS [1] leading to new insights into non equilibrium surface dynamics.

In this contribution we will report on experimental results of (a) on the surface dynamics of colloidal assemblies and (b) capillary wave dynamics of complex fluids covered by lipid monolayer.

(a) Surface XPCS has been used to measure the slow dynamics of gold particles on polymer films [1]. Above the glass transition of the polymer film the gold particles were found to slowly coalesce. This non equilibrium process is accompanied by a superdiffusive motion of the particles typical for jammed systems in a non-equilibrium system. We investigated also the dynamics of particles at the surface of a colloidal suspension and compare the dynamics to the corresponding bulk behaviour as measured with dynamic light scattering.

(b) We investigated the surface dynamics of DPPC monolayers adsorbed on sol subphases containing nanometer sized mineral particles. Capillary wave spectra have been measured upon compression of the DPPC monolayer revealing a dramatic change of the viscoelastic properties of the layer. This is attributed to the formation of a solid like DPPC layer which is connected to mineral particles present in the sol phase.

[1] S. Streit, C. Gutt, V. Chamard, A. Robert, M. Sprung, M. Tolan, Phys.Rev.Lett. 98, 047801 (2007)

Coherent diffraction patterns of a Spin Density Wave

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We report on coherent X-ray magnetic scattering studies of the spin density wave antiferromagnetism of pure chromium. Non resonant coherent X-ray scattering was used below the K absorption edge. The observed diffraction patterns give evidence for the presence of phase shifts of the magnetic order, embedded in the bulk.

Anomalous Surface Dynamics in Supported Polystyrene Films Near the Glass Transition

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The reduction of the glass transition temperature (T_g) in thin supported polymer films is of great interest. One proposed explanation is that close to the surface region there exists a thin layer with very low viscosity. In order to testify this possibility, we applied X-ray photon correlation spectroscopy (XPCS) to measure surface dynamics of polymer films. The samples were polystyrene (PS) films on hydrophobic silicon substrates with various molecular weights ($M_w = 11-900$ kg/mol). The film thickness ranges from 40 nm to 320 nm. These samples were annealed at 170°C in high vacuum for approximately 24 hours. The XPCS measurements performed at the beamline 8-ID-I in the Advanced Photon Source, Argonne.

In the wave vector range 10^{-3} - 10^{-1} nm⁻¹, at just above T_g , we have found a relaxation mode of the surface fluctuations at least 100 times faster (for $M_w = 129k$) than the capillary wave theory predicts. Surprisingly, this mode does not show much molecular weight dependence. When the temperature increases far above T_g (>150C), the surface relaxation becomes normal, as predicted by capillary wave theory [1]. Using the theory developed [2] in the previous work, the results cannot be explained with a model having bilayer of a thin low viscous layer on top. The failure of fitting of the bilayer model clearly indicates that, within the current experimental resolution, the possibility of the existence of a thin less viscous layer near the surface can be excluded. Hence, in our current study, the anomalous surface dynamics at close to T_g , faster than predicted with a bulk viscosity, is not a result of the surface phenomena, and must arise from within the film.

[1] H. Kim, et al., Phys. Rev. Lett. **90**, 068302 (2003).

[2] Z. Jiang, et.al., Phys. Rev. E **.74**, 011603 (2006).

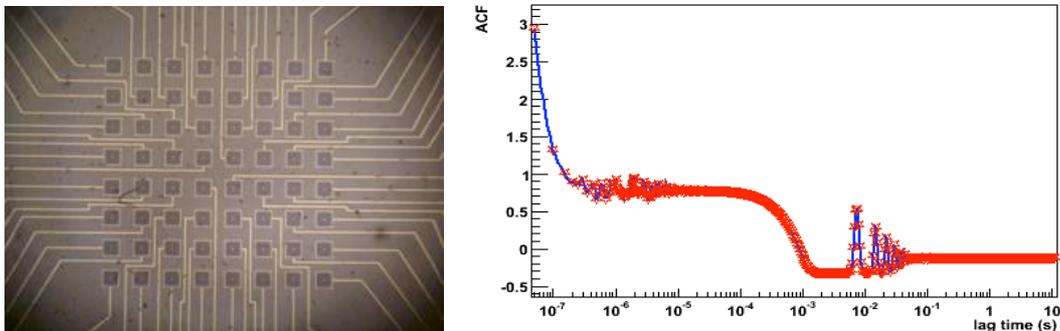
A fast 2D detector for X-Ray Photon Correlation Spectroscopy

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A fast (MHz) 2D X-ray detector will greatly improve our ability to probe weakly scattering, rapidly fluctuating soft matter systems with X-ray photon correlation spectroscopy (XPCS). XPCS probes the dynamics of a material by analyzing the temporal correlations among photons scattered by the material. Visible light PCS has been a valuable technique for studying the long wavelength hydrodynamics of fluids, including simple liquids, liquid mixtures, liquid crystals, polymers, and colloids. However, visible PCS cannot probe the short wavelength dynamics of materials or opaque materials. The new field of XPCS offers an unprecedented opportunity to extend the range of length scales over which a material's low frequency (10^1 to 10^6 Hz) dynamics can be probed down to interatomic spacings.

A two-dimensional detection system would increase the efficiency of such experiments by a few orders of magnitude, however currently available 2D detectors cannot provide frame rates of considerably more than 10-100 Hz. We are developing a 20 MHz frame rate, 2D X-ray detector using an 8x8 array of Geiger mode avalanche photodiodes (GMPDs), shown in Fig. 1. The combination of this array with a scintillation crystal, that partly converts the X-ray energy into visible photons, is sensitive to X-rays with energies from a few keV to tens of keV.



Left: 8x8 GMPD array ($100 \times 100 \mu\text{m}^2$ pixels). Right: First cross correlation measurement with this detector. The harmonic structure near 7 ms was induced by a beam chopping fan wheel.

This detector, the readout scheme, and the first auto-correlation and cross-correlation measurements will be the focus of this presentation.