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Non-diffusive dynamics in a colloidal glass: Aging versus rejuvenation

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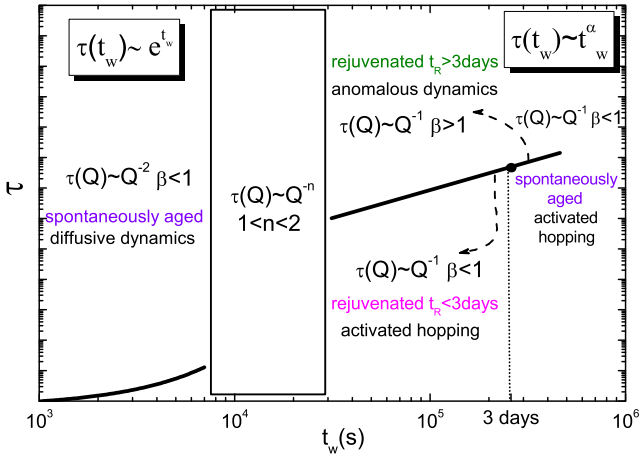
HIGHLIGHTS

- X-ray photon correlation measurements in Laponite glassy samples.
- Comparison between spontaneously aged and rejuvenated samples.
- Compressed and stretched behaviuors of the intensity correlation functions.
- Non-diffusive dynamics of the particles.

GRAPHICAL ABSTRACT



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ABSTRACT

The microscopic dynamics of spontaneously aged and rejuvenated glassy Laponite is investigated through X-ray photon correlation spectroscopy. Two different behaviours of the intensity autocorrelation functions are observed depending on the history of the sample: stretched for spontaneously aged samples and samples rejuvenated from a Wigner glass and compressed, typical of anomalous dynamics, for samples rejuvenated from a DHOC glass. The relaxation time behaviour in the three cases indicates a non-diffusive dynamics of the particles. The present system offers therefore an overview of various dynamical behaviours previously observed individually in several systems and the possibility to pass from one to the other choosing ad hoc the time parameter.

1. Introduction

Soft matter plays nowadays a central role in the development of advanced materials thanks to the variety of physical states which offers including liquids, foams, gels, glasses. Its interest arises from the ability that its microscopic constituents have to rearrange and form mesoscopic structures. It is challenging therefore to predict as much as possible these interesting behaviours from the microscopic to the mesoscopic scales in order to be able to manipulate and consequently control the macroscopic behaviour of such materials. To this purpose the experimental and theoretical study of the structure and dynamics of these systems is essential.

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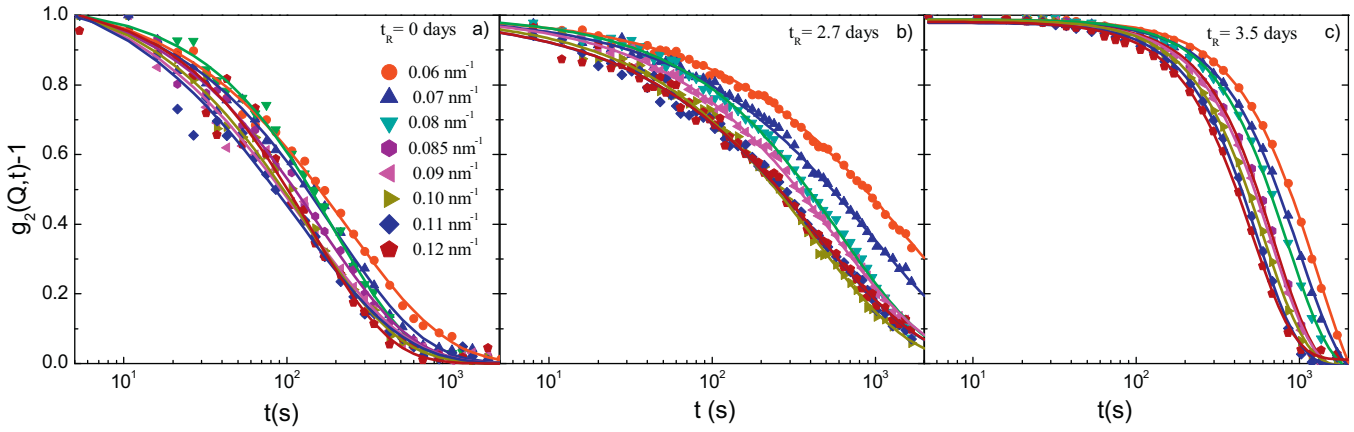

Fig. 1. Normalized intensity autocorrelation functions of an aqueous Laponite suspension at concentration C_w=3.0 % (a) for a spontaneously aged sample with t_R = 0 days ($t_w = 5.67 \times 10^4$ s), (b) for a rejuvenated sample with t_R = 2.7 days ($t_w = 1.84 \times 10^4$ s) and (c) for a rejuvenated sample with t_R = 3.5 days ($t_w = 1.97 \times 10^4$ s) and at different Q values obtained through XPCS (symbols). The solid lines represent the best fits performed using Eq. (1).

Studying the static and dynamic structure factors is the most direct way to access information on the characteristic length (dimensions) and time (relaxation times) scales typical of these systems. Their dynamical behaviour is characterized by the presence of relaxation processes [1] associated for example to the interactions between a particle and the cage of its nearest neighbors or related to the structural rearrangements of the particles. The signature of a relaxation process is a decay of the dynamic structure factor described, over a wide time window, by the Kohlrausch-Williams-Watts expression $f(Q,t) \sim exp[-(t/\tau)^{\beta}]$ where τ is an "effective" relaxation time and β measures the distribution of relaxation times. Usually in soft materials an exponent $\beta < 1$ is found, this behaviour is referred to as "stretched behaviour". On the contrary in the last decade an unusual behaviour characterized by a β exponent $\beta > 1$ has been observed and referred to as "compressed behaviour" [2–16]. These two peculiar trends correspond to different microscopic dynamics of the particles.

Here we present an overview of the dynamical behaviour of a prototype charged colloidal system, aqueous Laponite suspensions, characterized by an aging dynamics. Once dispersed in water Laponite ages towards different arrested states like equilibrium gel and glass depending on ionic strength and particle concentrations [17]. In this work we focus on the glass state at concentration $C_w = 3.0$ % largely investigated experimentally through different complementary techniques such as Dynamic Light Scattering [3,18,5,19], Rheology [20–22], Small Angle X-ray Scattering [23–25], Small Angle Neutron Scattering [26], X-ray Photon Correlation Spectroscopy [4,13,15,27,28] and Neutron Spin Echo [27]. The waiting time (t_w) (time scale) and Q (spatial scale) dependence of the structural relaxation process of spontaneously aged and rejuvenated samples are studied. Both stretched and compressed behaviours of the intensity autocorrelation functions are found and discussed in relation to previous studies and on the light of the presence of attractive and repulsive microscopic interactions versus the application of external stresses. The different dynamical behaviours observed here in a single system are general features individually observed in several systems as molecular liquids [29,30], colloidal hard spheres [31], colloids [2,10], clays [3–5,13,15], metallic glasses [11], polymers [6,7,9,14], supercooled liquids [8].

2. Materials and methods

2.1. Materials

Laponite is a synthetic clay that, when dispersed in water, forms a charged colloidal suspension of platelets with 25 nm diameter and 0.9 nm thickness and inhomogeneous charge distribution, negative on the faces and positive on the rims. The platelets are usually considered monodisperse in size but a small polydispersity has been reported by different authors [32,33].

Aqueous dispersions of Laponite RD with weight concentrations $C_w = 3.0$ % were prepared in a glovebox under N_2 flux to avoid contact with air and prevent CO_2 degradation [34]. Laponite powder, manufactured by Laporte Ltd., was dispersed in pure deionized water, stirred vigorously for 30 min and filtered soon thereafter through a 0.45 μm pore size Millipore filter in bottles sealed in the glovebox. A part of the stock solution was directly filtered in glass capillaries of 2 mm diameter for the experiments. These were later referred to as "spontaneously aged" samples. The origin of the waiting time ($t_w = 0$) determines the age of the sample and for the spontaneously aged samples it is the time at which the suspension is filtered. Rejuvenated samples were prepared starting from the stock solution that had rested in the bottles some time t_R (rejuvenation time) since filtration and injected into the capillary through a syringe, hence introducing a huge shear field (shear rejuvenation). The age t_w of a rejuvenated sample is counted from t_R. Even if rejuvenation by a shear field corresponds to returning the sample to earlier aging times, however the process never rewinds the sample to the original as prepared one.

2.2. Measurements

The samples were characterized by X-ray Photon Correlation Spectroscopy (XPCS) [35,36] at beamline ID10A of the European Synchrotron Radiation Facility (ESRF) in Grenoble. For the measurements a partially coherent and monochromatic X-ray beam with a photon energy of 8 keV was employed. Long series of scattering images were recorded by a charged coupled device (CCD) placed in the forward scattering direction. The images were post processed following the multi-speckle XPCS approach [35] to get access to the dynamics of the samples. Ensemble averaged intensity autocorrelation functions $g_2(Q,t) = \langle \frac{\langle I(Q,t_0) I(Q,t_0+t) \rangle_p}{\langle I(Q,t_0) \rangle_p \langle I(Q,t_0+t) \rangle_p} \rangle_{t_0}$ were calculated using a standard multi-tau algorithm. Here, $\langle \ldots \rangle_p$ indicates averaging over pixels of the detector mapping onto a single value of the momentum transfer (Q) while $\langle \ldots \rangle_{t_0}$ indicates temporal averaging over t_0.

3. Results and discussion

The XPCS intensity autocorrelation functions of aqueous Laponite suspensions as a function of Q are shown in Fig. 1 for