X-ray beam induced dynamics in silica-based glasses

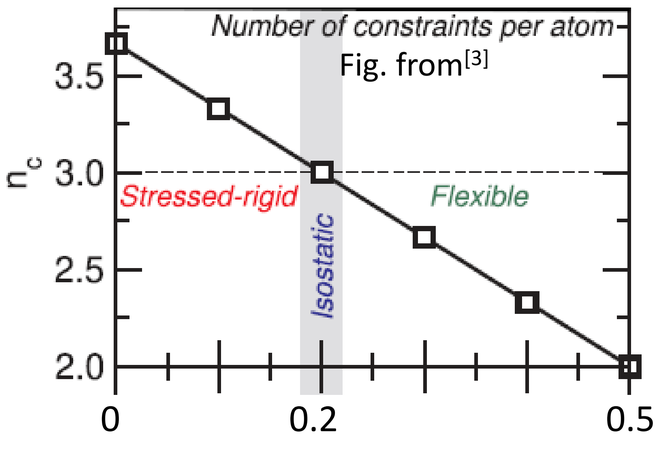
E. Alfinelli[1], F. Caporaletti[2], R. Dal Maschio[3] , A. Martinelli[1], G. Monaco[4], B. Ruta[5, 6], M. Sprung[7], M. Zanatta[1], G. Baldi[1]

[1]Physics Department, Trento University, I-38123, Povo, Trento, Italy. [2]Faculty of Science, University of Amsterdam, 1090 GL, Amsterdam, The Netherlands. [3]Department of Industrial Engineering, , Trento University, I-38123, Povo, Trento, Italy. [4]Physics and Astronomy Department, I-35122, University of Padova, Italy. [5]ESRF- The European Synchrotron, F-38043, Grenoble, France. [6]Institut Lumière Matière, UMR5306 Université Lyon 1–CNRS, Université de Lyon, 69622, Villeurbanne Cedex, France. [7]Hasylab at DESY, Notkestr. 85, D-22607 Hamburg, Germany

INTRODUCTION

X-ray Photon Correlation Spectroscopy (XPCS) is one of the most promising techniques to study the dynamical properties of materials at the nanometer scale up to hundred of seconds, such as colloidal samples and structural glasses. However, recent studies [1],[2] have shown that the X-ray beam induces the sample dynamics, pumping and probing at the same time the atomic displacements: this phenomenon is yet under and opens the possibility to investigate new properties of glasses. We have investigated this effect in a set of silica-based samples as a function of both the scattering vector and the dose.

EXPERIMENTAL



The samples are pure silica glass and two sodium silicate glasses ($(Na_2O)_x(SiO_2)_{1-x}$) at different sodium concentration (x=20%, x=40%). Samples are prepared via the melt-quenching technique in which the soda and silica powder are melted into a furnace and then quenched in air. Samples characterization was carried out via Raman spectroscopy in Trento laboratories.

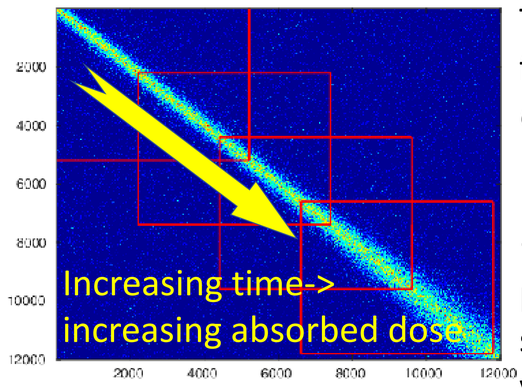
-> *Increasing sodium concentration*

The investigation of the dynamics has been performed with XPCS at the beamline P10 of the PETRA III synchrotron in Hamburg.

Characteristic	Value
Energy	8.25 keV (0.15 nm)
Photon Flux	$\sim 10^{11}$ ph/s
Beam Spot	(2x3) μm^2
$r_{speckle}$	92 μm
Detector	Eiger X4M (2070x2167)
Pixels size	(75x75) μm^2
Temporal width	3 μs - 10^3 s

2D photon counting detector.
Multi-Speckle approach

DATA ANALYSIS

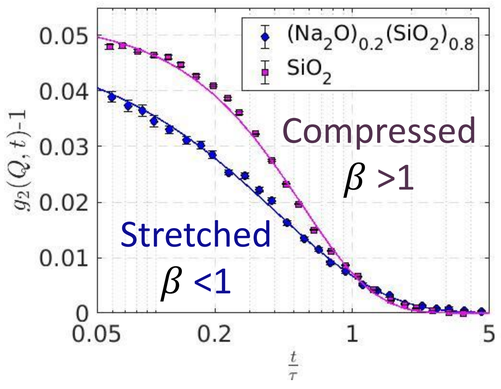


Two times correlation function(TTCF).

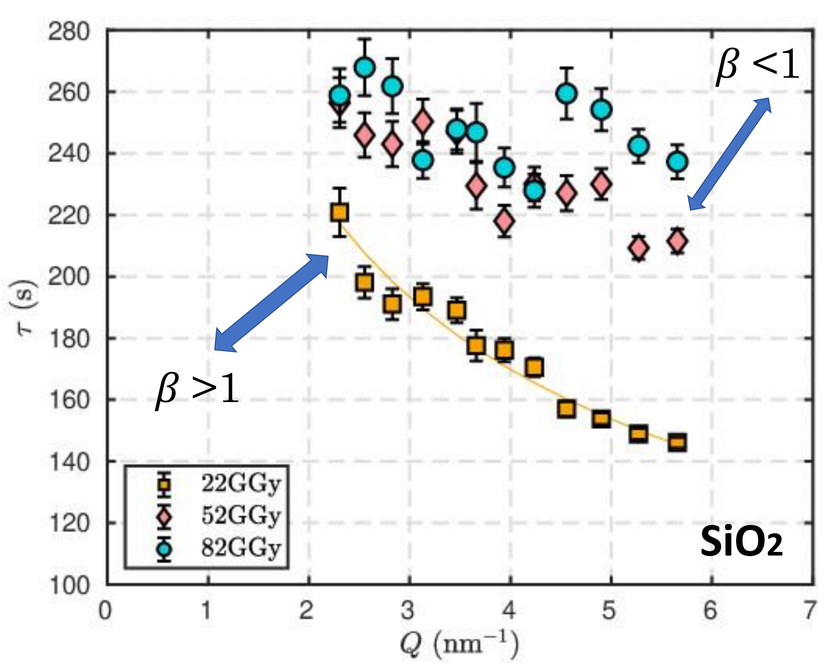
$G(q, t_1, t_2) = \frac{<I_p(t_1)I_p(t_2)>_p}{<I_p(t_1)>_p<I_p(t_2)>_p}$, with $< \cdots >_p$ the average performed over pixels subtending the same q vector (Product of the intensities at different times).

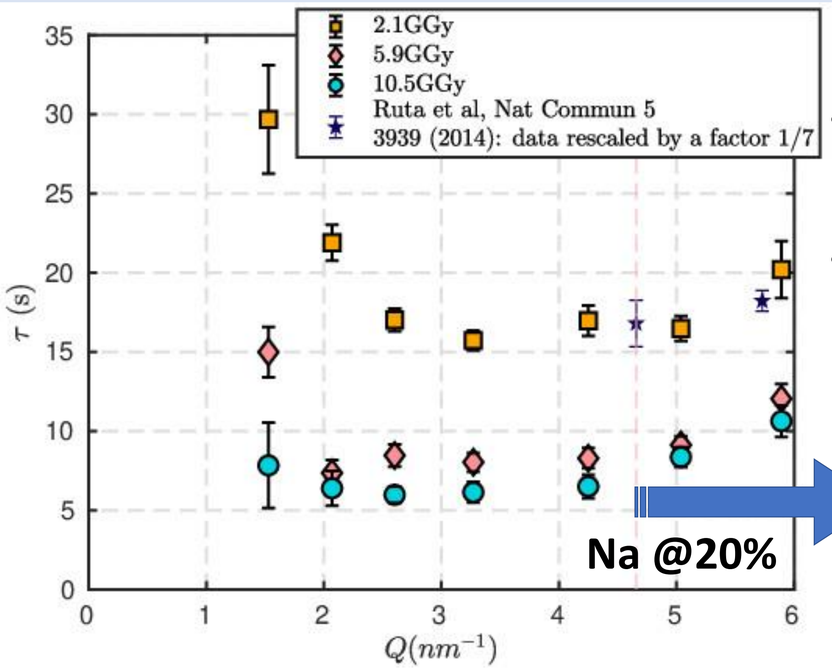
The sample dynamics is recovered by performing time average inside narrow temporal windows (in red). The resulting autocorrelation function is then fitted with the KWW model.

$$g_2(Q,t) = 1 + C(Q)e^{-2\left(\frac{t}{\tau}\right)^{\beta}}$$



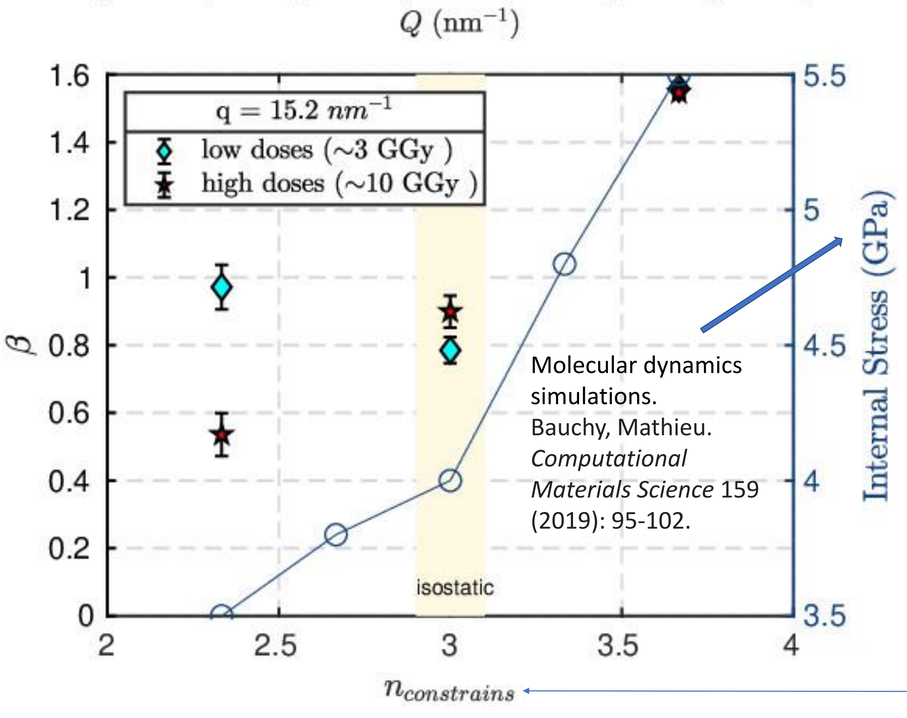
RESULTS



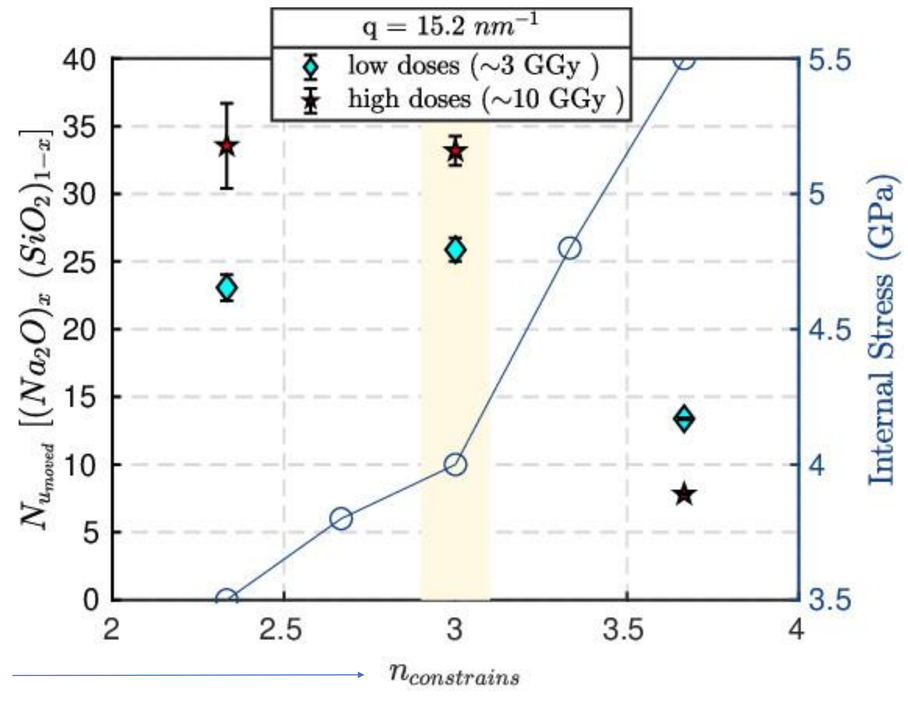


- Loss of the Q (scattering vector) dependence at high doses (Gy) rates
- Investigation of a new Q range by closing the entire optical path in vacuum tube (mimic of SAXS experiments)

Limit in Q of literature data



Stressed silica glass presents a beta value almost independent of the absorbed dose: presence of higher internal stresses. Simultaneously the atoms moved after radiolysis phenomenon are lower with respect to the other samples.



Calculated from Fig. in EXPERIMENTAL

CONCLUSIONS

- The induced dynamics is modified and the beta parameter decreases when the absorbed dose exceeds a threshold value.
- Beta follows the local stress from MD simulations.
- The addition of sodium to pure SiO_2 modifies the Q-depedence of the observed dynamics.
- For high dose values, the de-correlation time looses any Q-dependence @ low Q's and beta decreases below one, as in the supercooled liquid case.

[1]B. Ruta et al, Sci. Rep, 7(1):1–8, 2017, [2]B. Ruta et al, Nat Commun 5, 3939 (2014), [3]Bauchy, Mathieu. Computational Materials Science 159 (2019): 95-102.