Study of the vibrational spectrum in glassy systems

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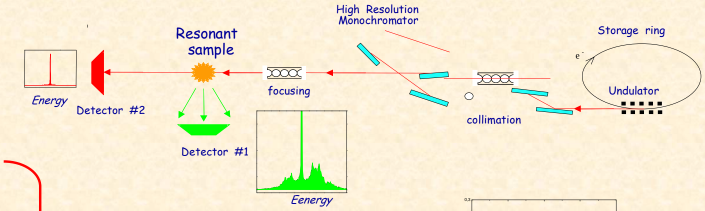
Introduction

The aim of the present study is to investigate the dynamical behaviour of the glassy systems, in particular we try to understand the microscopic nature of an anomaly, the Boson Peak (BP), present in their vibrational spectra at low energy.

NIS Experimental layaout



The technique

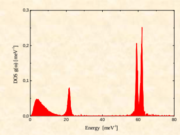
We probe the dynamics of glassy phase thanks to the use of the nuclear inelastic scattering (NIS) [1]. This technique exploiting the low energy nuclear transition of the Fe^{57} isotope, it is able to collect the purely incoherent signal emerging from the sample. Starting from the NIS spectrum it is possible to derive without any model the vibrational spectrum g(w) experienced from the Fe atoms present in the glass.

$$W(E) = f_{LM}\left(\delta(E) + \sum S_n(E)\right)$$

$$S_n(E) = \frac{1}{n}\int S_n(E')S_{n-1}(E-E')dE$$

$$S_1(E) = \frac{E_R\, g(E)}{E(1-e^{-\beta E})}$$

$$g(E) = \frac{V}{(2\pi)^3}\sum_j \int dq\, \delta(E - \hbar\omega_j(q))$$



The DOS

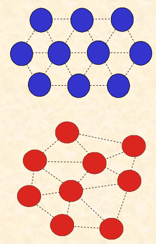
The dynamics of a system can be well characterized by its vibrational spectrum DOS (the vibrational density of state). In particular an universal feature in the DOS seem identify the disordered structure of a glass: the Boson Peak [2]. This anomaly is an excess of vibrational modes in the reduced density of state (RDOS) $g(w)/w^2$ relative to that expected for the correspondent crystalline phase and predicted by the Debye model.

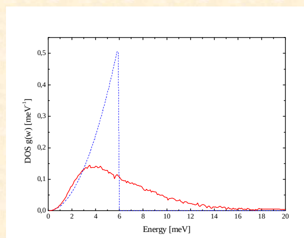
Ordered phase.

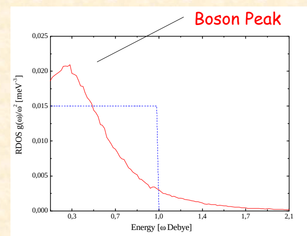
Debye model.

$$g_D(E) = \frac{3}{\omega_D^3}\omega^2$$

Disordered phase.







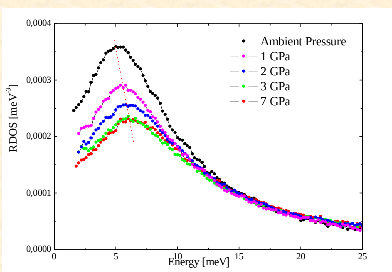


Fig. 1 RDOS of $Na_2FeSi_3O_8$ at different P

Experimental investigation RDOS and S(Q) vs P

Looking to the pressure dependence of the DOS in a densified silica glass $Na_2Fe^{57}Si_3O_8$, we observe that the BP has a well defined tendency. As shown in Fig. 1 increasing the pressure the BP decreases in intensity and shift to higher energies. The pressure produce also an evident effect on the structure of the system, in particular the first peak of the static structure factor shows an evident change in shape from 0-7 Gpa Fig.2.

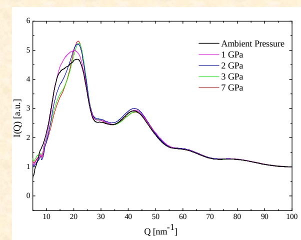
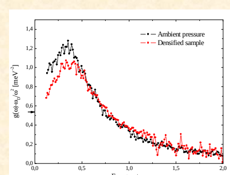


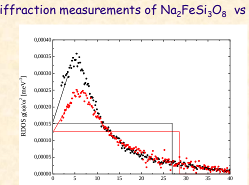
Fig. 2 diffraction measurements of $Na_2FeSi_3O_8$ vs P

- Why the pressure dependence of the BP ?
- What is the connection between the dynamics of the glass and its mycroscopic structure?
- The evolution of the BP vs P it is simply driven by the increase of the elastic constants in the glass under pressure?

In order to understand if the evolution of the BP vs P is simply associated to the change of the corresponding Debye frequency wd vs P, we renormaliz the RDOS by the values of wd estimated from measurements of light scattering,

The normalized spectra don't converge to a unique master curve!!





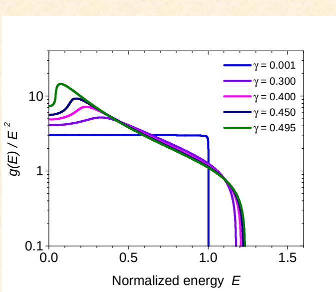
A theoretical scheme

A possible explanation of the observed dependence of BP vs P, can be found in a theoretical scheme that associate the presence of the BP to the dynamical disorder of the system [3] . Molecular dynamics simulations show in simple models that the atoms experience fluctuations Δk in their force constants K. The BP increase in intensity increasing the fluctuations Δk.

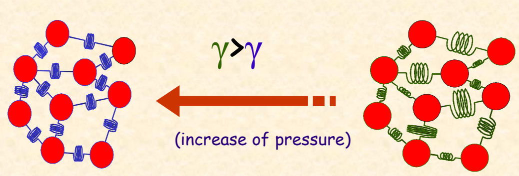
Dynamical disorder = disorder in the elastic constants K

k_0 $K = k_0 + \Delta k$ $\gamma = \frac{\langle \Delta k^2 \rangle}{\langle k \rangle}$

γ, homogeneity parameter of the elastic structure of the system



decrease of the BP ← Increase of the elastic order (increase of pressure)



$\gamma > \gamma$

(increase of pressure)

Conclusions

Our experimental investigations of the DOS in densified glasses give us the opportunity to investigate the connections between the dynamic of the system and its structure. Moreover the present study represent a starting point to test a theoretical argument that relate the BP to the non homogeneity of the elastic structure of a system , in this scheme the pressure appear as a pertinent tool to change the dynamical disorder of the glass.

References

[1] A Chumakov et al. Hyper. Inter. **121-123** 781 (1999)
[2] A. P. Sokolov et al. Phys. Rev. Lett. **78** 2405-2408 (1997)
[3] W. Shirmacher et al. Phys. Rev. Lett. **81** 136 (1998)