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NUCLEAR INELASTIC SCATTERING IN DENSIFIED AND HYPERQUENCHED GLASSES

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In the last years the dynamics of the glassy phase has been extensively investigated: it reveals aspects that cannot be directly associated neither with the lack of order (relative to the crystalline phase), neither to the loss of mobility (relative to the liquid phase).

With the present study we try to clarify this peculiarity of glasses focusing our interest on the investigation of the so-called Boson Peak (BP) [1]. The BP appears in the vibrational spectra of basically all glasses as an excess of low-frequency modes compared to the Debye model expectations. The universal character of the BP for glasses leads us to address some basic questions about its origin: Is the BP associated with the structural disorder of the system? Is the non-equilibrium nature of a glass responsible for this phenomenology?

We investigate the connection between vibrational properties, structure and energy landscape description of a glassy system. To fulfill this task, we probe how the vibrational spectrum of a glass evolves with density and as a function of its thermal history using nuclear inelastic scattering (NIS) [2].

First, we measured the vibrational spectra of densified glasses, i.e., systems that remain at elevated density after an external pressure (0-7 GPa) is released. We observed that the density has a pronounced effect on the BP: On increasing the density, the BP moves toward higher energies and decreases in intensity (Fig.1, left), with an evident correlation between position and height of the peak.

Second, in order to probe how the non-equilibrium nature of a glass can control its dynamics, we measured the vibrational spectra for hyperquenched materials. In fact, by fast freezing a liquid with various cooling rates, it is possible to obtain glasses with structure and potential energy corresponding to various *fictive temperatures* T_f (the temperatures where the system falls out of equilibrium during cooling) [3]. Figure 1 (right) shows that the intensity of the BP increases for samples obtained with higher cooling rates (Fig. 1). This result has been theoretically predicted [4], but till now only partially experimentally verified [5].

The dependence of the BP on density and cooling rate shows that its phenomenology is controlled by both the structure and the energy landscape of the system.

These experimental observations strongly indicate that in glassy systems dynamics, structure and potential energy are tightly interconnected.

Furthermore, our measurements can help to clarify the validity of theories proposed in the last years to explain the nature of the BP.

[1] see, for instance, A. P. Sokolov *et al.*, Phys. Rev. Lett. **78** (1997) 2405.
[2] see, for instance, A. Chumakov *et al.*, Hyperfine interactions **123-124** (1999) 781.
[3] C. A. Angell *et al.*, J. Phys.: Cond. Matt. **15** (2003) S1051.
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[5] E. Duval *et al.*, Europhys. Lett. **63** (2003) 778.

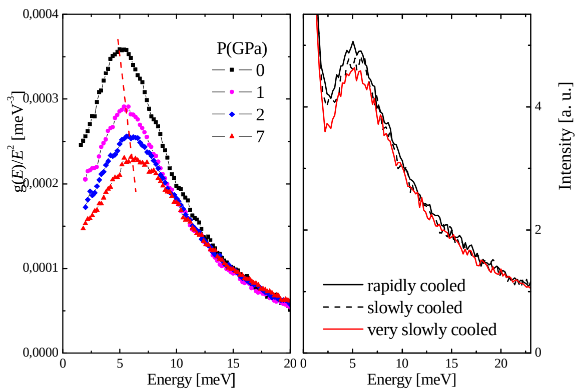

Figure 1. Left: Reduced density of states RVDOS $g(E)/E^2$ of $Na_2FeSi_3O_8$ for different densities obtained pressurizing the sample at 0, 1, 2, and 7 GPa. Right: Energy spectra of NIS for $Na_2O/FeO/CaO/SiO_2$ glasses prepared with various quenching rates.