



	Experiment title: Separation of propagating and localized vibrational modes in glasses	Experiment number: HS-1931
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Report:

We attempted to separate collective extended motions from local vibrations in dynamics of glasses and to find out which ones of them cause the Boson peak (an excess of low-energy states relative to a crystal). We found that such separation indeed can be achieved with nuclear inelastic scattering (NIS) [1] by studying dynamics of glasses with resonance probes, and that the Boson peak in studied samples belongs to collective extended motions.

Toluene and ethyl benzene glasses were investigated in ~20-100 K temperature range. The dynamics of glassy matrix were studied by acquiring information on motions of a probe ferrocene molecule, in particular, those of a resonance ^{57}Fe nucleus at a center of the probe. Besides measurements in glassy states, we performed measurements in crystalline phases of both toluene and ethyl benzene.

Fig. 1 shows density of vibrational states $g(E)$ for toluene glass at 22 K. Sharp peaks at 22, 59 and 62 meV arise from motions of the central Fe atom within the ferrocene probe. They are identical to those in crystalline ferrocene. Vibrational states below ~17 meV describe motion of the entire probe grasped by sound-like collective extended motions of glassy toluene matrix. Density of states of these motions is quite different from lattice vibrations in a crystalline ferrocene (Fig.1).

Up to ~2.5 meV, our data on density of states in glassy toluene follow neutron data [2]. At higher energy they show less number of states. In particular, they do not show any noticeable contributions from known local modes of methyl group rotation (~6-12 meV), methyl group torsion (27 meV) and some other local intermolecular modes of toluene at ~42, 48, and 57 meV observed in neutron data. Thus, an application of resonance probes does allow one to measure selectively collective extended motions of glass matrix and to ignore local vibrations of molecular fragments.

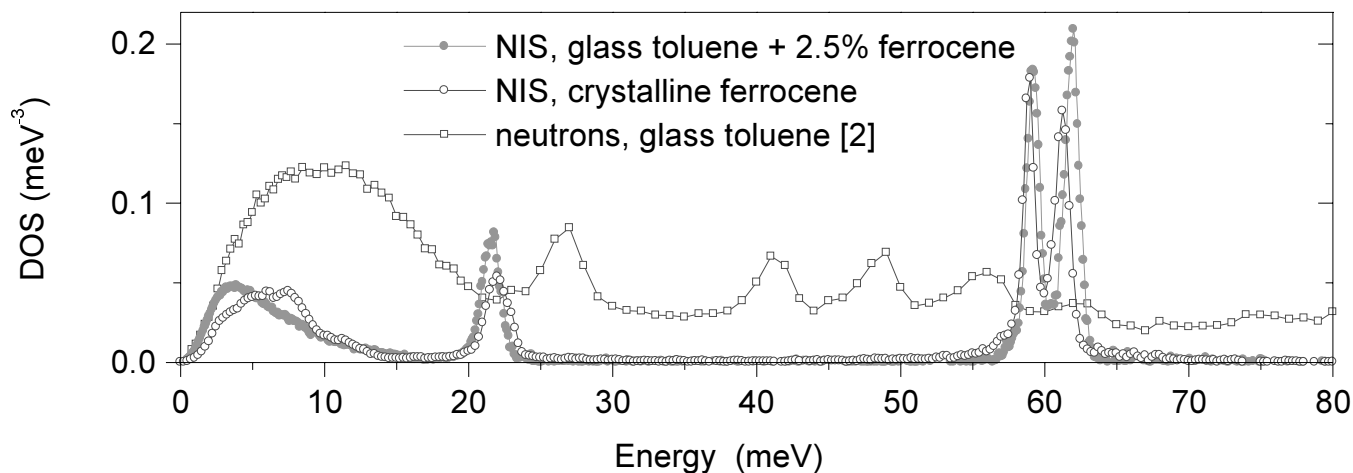


Fig. 1. Density of vibrational states for toluene glass measured with probe ferrocene molecules (●), crystalline ferrocene (○), and neutron data [2] from toluene glass (□).

At low temperature (22 K), reduced density of states $g(E)/E^2$ in toluene shows the Boson peak at ~ 1.5 meV, in accordance with light scattering data [3]. At higher temperatures, shape of the Boson peak becomes flatter. Closer to glass transition ($T=120$ K), the Boson peak disappears under growing contribution of low-energy states. Density of states of both glasses showed an excess of low-energy states relative to their crystalline phases (Fig. 3).

In summary, we have observed the Boson peak in toluene and ethyl benzene glasses. Energy position of the peak and its temperature evolutions are in agreement with previously reported data. On the other hand, comparison to neutron scattering (Fig. 1) shows that our data are sensitive exclusively to collective extended but not to local vibrations. On this basis we conclude that the Boson peak in studied glasses belongs to collective extended motions.

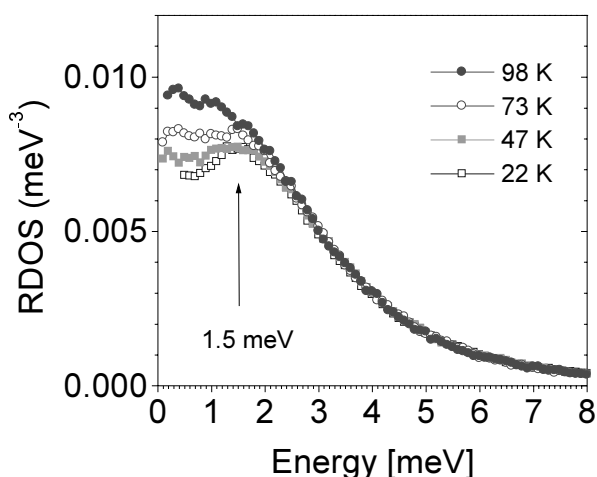


Fig. 2. Reduced density of states $g(E)/E^2$ of toluene at various temperatures.

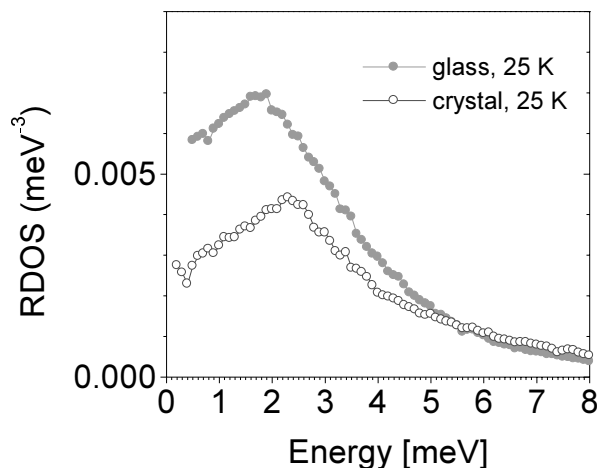


Fig. 3. Reduced density of states $g(E)/E^2$ of glassy and crystalline ethyl benzene.

References:

1. see, e.g., A.Chumakov and W.Sturhahn, *Hyp.Interact.* **123/124** (1999) 781.
2. I.Tshukushi, O.Yamamuro, K.Yamamoto *et al*, *J.Phys.Chem.Solids* **60** (1999) 1541.
3. J.Wiedersich, N.V.Surovtsev and E.Rössler, *J.Chem.Phys.* **113** (2000) 1143.