ESRF	**Experiment title:** Does the "boson peak" exist?	**Experiment number**: HS-1323
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Report:

We applied nuclear inelastic scattering to investigate the vibrational dynamics of glassy dibutyl phthalate using dissolved ferrocene probe molecules enriched in resonant isotope ^{57}Fe. We wanted to clarify if a low-energy excess of vibrational modes in glasses (the boson peak) can be explained by multi-phonon contributions to the density of states. Measurements were done for 7 temperatures in the range 3-200 K.

We observed the boson peak and found that it cannot be attributed to multi-phonon contributions. Fig. 1 shows raw experimental data, derived density of states $g(E)$ and reduced density of states $g(E)/E^2$ at 50 K. The "boson peak" shows up at 2 meV as a deviation of the reduced density of states from the flat Debye approximation.

Further analysis showed that the application of resonant probe molecules is an efficient approach and provides important information on the nature of the boson peak. A neutral molecule immersed in a neutral glass matrix is sensitive only to center-of-mass vibrations and not to internal modes of glass molecules.

The density of states (Fig. 1) consists of vibrations of the glassy matrix (below 20 meV) and 3 Einstein-like modes of ferrocene (at 21, 58, and 61 meV). Vibrational modes of ferrocene and a contribution of iron atoms to these modes are well known. This allowed us to calculate the number of modes of glass dynamics below 20 meV. The result is 3.08±0.1. Therefore the vibrations of the glass matrix are composed solely of 3 acoustic-like modes, whereas all localized intra-molecular vibrations and rotations of glass are excluded. The center-of-mass modes do show the boson peak at 2 meV, the same position as obtained by another group with inelastic neutron scattering.

The short wavelength of the applied radiation (0.86 Å) provides high sensitivity of the data to atomic vibrations. At T>130 K the multi-phonon contributions are dominant in the raw spectra. Calculations of multi-phonon contribution within harmonic approximation gives a test to which extent the vibrations are harmonic. Fig. 2 shows the raw data, density of states, and reduced density of states for various temperatures. The raw spectra vary much with temperature. Above 100 K this variation is caused mainly by multi-phonon absorption. Despite of that, the derived densities of state are identical within 3-175 K temperature range. This indicates that the vibrations stay harmonic up to the vibration amplitude of 0.2 Å.

These findings draw the conclusion that the "boson peak" in glassy dibutyl phthalate is caused by harmonic center-of-mass vibrations of glass molecules.

Besides that, various hypothesis on the microscopic explanation of the boson peak can be tested in view how they can explain that the probe molecules "see" the peak. For instance, the suggestion on acoustic origin of the boson peak is consistent with the data, because the acoustic waves propagate through the entire media. Within the opposite hypotheses of localized vibrations of glassy clusters, our data give an upper limit of the cluster size: from the mean force constant (calculated from the raw spectra) and the boson peak frequency we estimate the largest cluster size as 2-3 glass molecules.

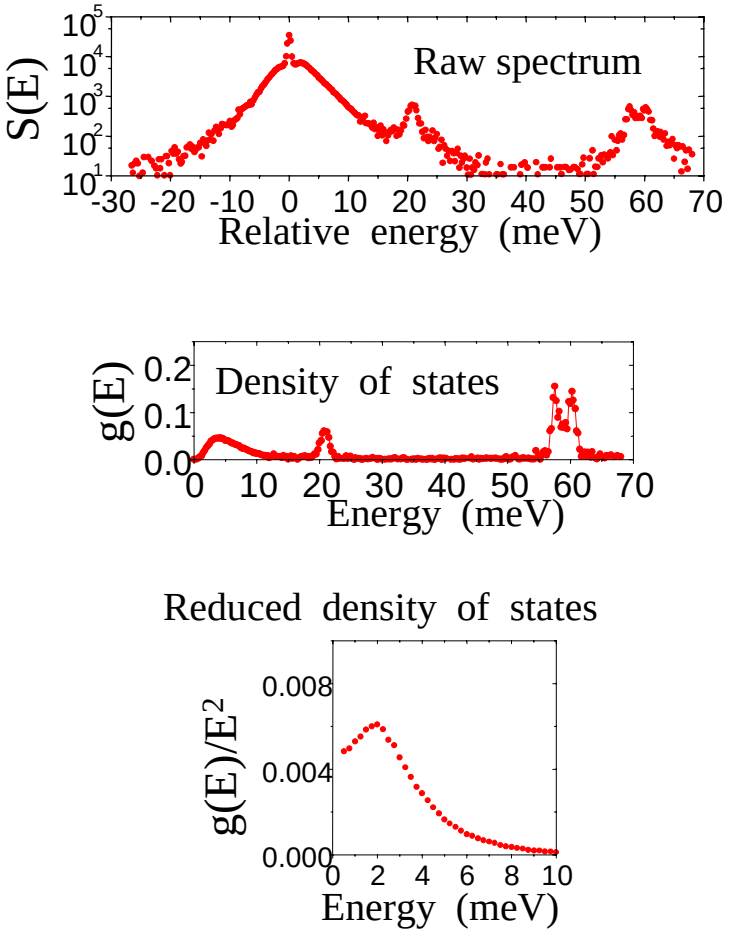

Figure 1

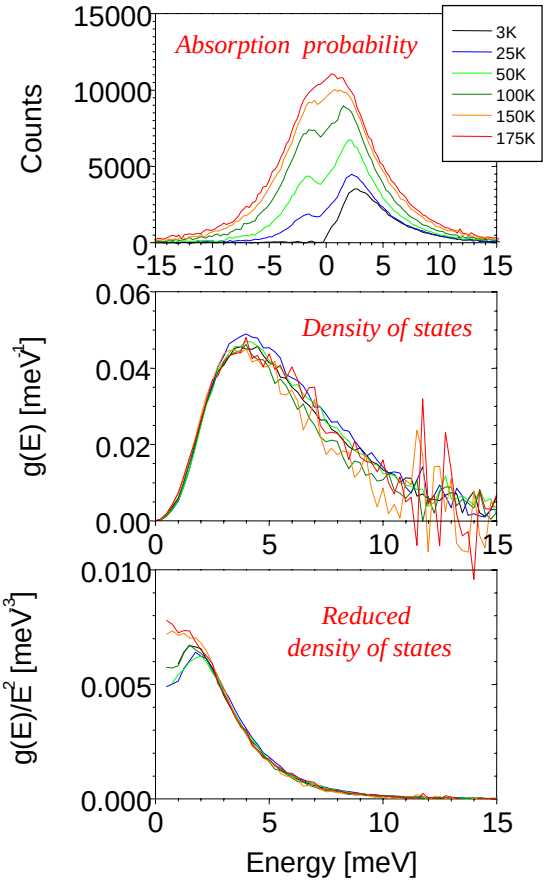

Figure 2