Glass dynamics as seen by nuclear inelastic scattering

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We investigated the fast dynamics of several molecular glasses in the temperature range from the glass transition down to ~10 K. The density of the vibrational states of methylbenzene, ethylbenzene, dibutylphthalate and glycerol was measured by nuclear inelastic scattering [1] using nuclear resonant probes dissolved in the glassy matrices. The nuclear probes were ferrocene molecules for the first three glasses, and Fe^{+2} ions in the case of glycerol. Due to the chosen experimental approach, the obtained data relate mainly to long-wavelength collective translational motions of the centers of mass of the glass molecules and are much less sensitive to local vibrations. For the first three glasses, for instance, the contributions of known local vibrations of methyl groups and other molecular fragments were completely excluded.

At low energies, the energy dependence of the density of vibrational states exhibits at low temperatures for all studied glasses the typical deviation from the quadratic Debye behavior known as the "Boson" peak. The energies of the observed Boson peaks agree with those available from neutron scattering data. At higher temperatures, the shape of the Boson peak revealed a known development: The density of states below the Boson peak grows with temperature and the peak becomes smoother. Finally, it disappears when the temperature approaches the glass transition temperature. On this basis, we conclude that the Boson peak in the studied glasses belongs mainly to long-wavelength collective translational motions of the centers of mass of the glass molecules.

At energies above the Boson peak, the density of states $g(E)$ exhibits for all studied glasses an universal temperature-independent behavior

$$g(E) \propto E^2 \exp(-E/E_0).$$

The characteristic energy E_0 correlates with the energy of the Boson peak. This behavior continues to energies as high as the data are free from a contribution of local vibrations, i.e., up to 17 meV for the first three glasses and up to 9 meV for glycerol. Comparing our results to available data on other glasses, we found a similar behavior for orthoterphenyl [2] and ZrTiCuNiBe and ZrAlNiCu metal glasses [3], although not explicitly pointed out by the authors. No similarity, however, was found for vitreous silica [4], where the Boson peak is known to originate from local librations of coupled SiO_4 tetrahedra.

References

[1] see, e.g., several contributions in E. Gerdau and H. de Waard, *Nuclear Resonant scattering of Synchrotron Radiation*, Hyperfine Interactions**, 123/124** (2002)

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