ESRF

Experiment title: Glass dynamics and scaling behaviour under pressure using quasielastic nuclear forward scattering		Experiment number: SC-773
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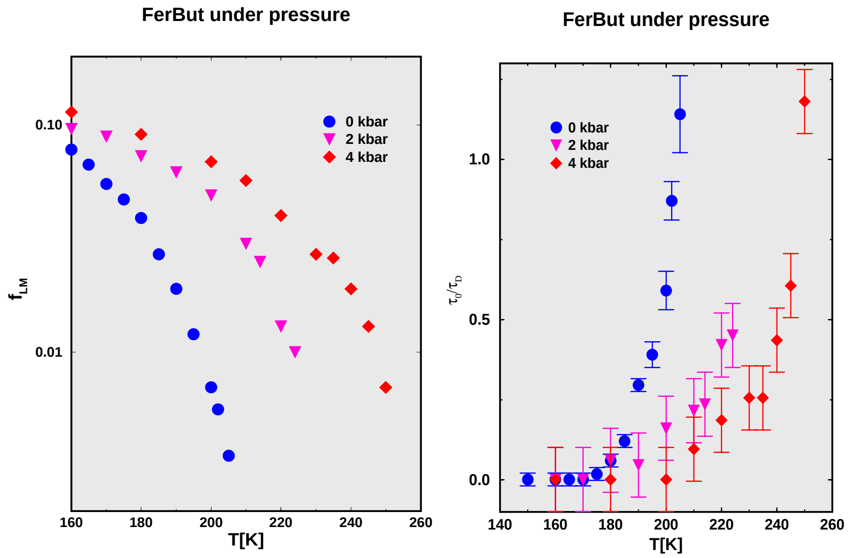
Report:

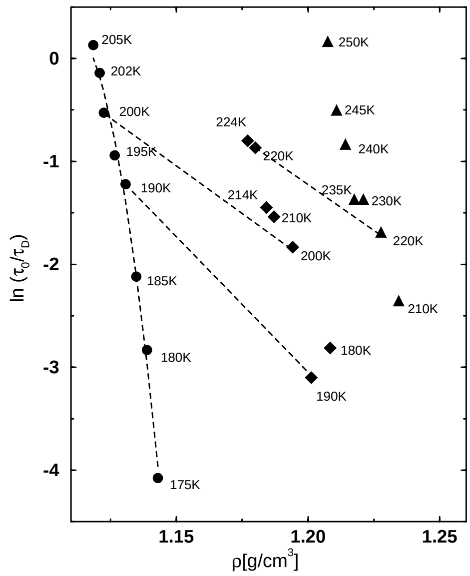
For practical reasons, combined studies of glass dynamics where both temperature and pressure are varied are rather scarce in literature. Such studies are nevertheless invaluable if one wishes to test the validity of various theories, such as mode-coupling theory [1,2] or the still largely popular free-volume model [3] for structural α-relaxation. Specific-heat spectroscopy of glassy ortho-terphenyl (OTP) at two different temperatures [4], for example, provided a strong evidence for the failure of free-volume models. The time window of specific-heat spectroscopy is limited to large relaxation times above $\sim 10^{-5}$ s; quasielastic nuclear forward scattering (QNFS) of synchrotron radiation [5] extends the accessible range down to $\sim 10^{-8}$ s. We have therefore applied the QNFS technique to dibutylphthalate doped with 5 atomic % ferrocene (enriched to 95.3 % in ^{57}Fe) at various pressures and temperatures.

The QNFS experiment was carried out at the beamline ID22N at the European Synchrotron Radiation Facility. The pressure apparatus (available at ID16) consisted of a closed-cycle cryostat with a specially designed cell having entrance and exit diamond windows of 1 mm thickness, respectively, a spindle and a Nova-Swiss pressure gauge. Forward count rates were in the range of 15 Hz (160 K/2 kbar; 180 K/4 kbar) down to 0.5 Hz (220 K/2 kbar; 250 K/4 kbar). The spectra, which showed a simple quantum beat structure owing to the quadrupole splitting of ferrocene, were subjected to a first preliminary fit assuming exponential diffusion; a more detailed data evaluation is under way.

The resulting Lamb-Mößbauer factors and diffusion times are shown in Fig. 1 (left and right side, respectively). We observe the expected stiffening of the "lattice" as well as a shift of the onset of relaxation to higher temperatures.



Lamb-Mößbauer factors (left) and diffusion times (right) for ferrocene/dibutylphthalate for three different pressures



Diffusion times for ferrocene/dibutylphthalate scaled versus density. Circles: atmospheric pressure, diamonds: 2 kbar, triangles: 4 kbar. The atmospheric-pressure data and some isotherms are provided with guides to the eye (dotted lines)

Using known V, P data and thermal expansion coefficients and applying the Tammann-Tait equation [6] to calculate the density, we obtain the plot of the diffusion times versus density shown in Fig. 2. We can conclude – in analogy to the study of OTP [4] (see Fig. 5 in this paper) – that the diffusion times do not show the same scaling behaviour, depending on whether density is changed by applying pressure or by cooling/heating. Thus the validity of the free-volume model seems to be disproved also in our case of high q.

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