ESRF	**Experiment title:** X-ray and neutron brillouin scattering from an amorphous polymer	**Experiment number:** HS-370
Beamline: ID16	**Date of Experiment:** from: 03 / 09 / 97 to: 11 / 09 / 97	**Date of Report:** 21 / 02 / 98
Shifts: 21	**Local contact(s):** Alain Mermet	Received at ESRF:

Names and affiliations of applicants (*indicates experimentalists) :

L. Comez , D. Fioretto*, L. Verdini* , Dip. Fisica, Univ. Perugia, via Pascoli, I-06100, Italy*
G. Ruocco, Dip. Fisica, Univ. Aquila, I-67100, Italy*
U. Buchenau, D.Richter*, A. Sokolov*, IFF/KFA, 52425, Germany*
F. Sette, ESRF Grenoble, France*
B. Frick, ILL Grenoble, France*

Report:

Inelastic X-ray measurements of polybutadiene were performed at 12 different temperatures in the range *50-320K.* At each temperature, 5 different Q values have been tipically detected, i.e. $1, 2, 4, 7, 10nm^{-1}$. To better reconstruct the dispersion curves, other sets of Qs were measured at two significative temperatures. In particular, 3,6,9,12, $15nm^{-1}$ were measured at 294 K and, in addition, 1.5,4.5,7.5,10.5 and $13.5nm^{-1}$ spectra were taken at T = 140K.
In order to extract the relevant spectroscopic features of the spectra, such as the frequency position Ω and the width Γ of the inelastic peaks, and the intensity of both elastic and inelastic peaks, a fitting procedure was used. It was based on the convolution of the experimentally determined resolution function with a model function. The central line was represented by a Lorentzian and the inelastic peaks by a DHO (damped harmonic oscillator).
Fig.1 shows the frequency position and the width of the inelastic peaks of the spectra taken at 140K for different values of the scattering vector. The linear behavior of Ω for Q < 5 indicates the propagating acoustic nature of the mode responsible for the inelastic scattering. The slope of the line gives the sound velocity v_{IXS}. A different Q-dependence is found for the width Γ which, for Q < 5 is well represented by a Q^2 law. For Q > 5 the value of Γ overtakes that of Ω, and acoustic-like excitations progressively loose their propagative nature.
In Fig.2 the temperature behavior of v_{IXS} is reported together with that obtained by Brillouin light scattering (BLS) and ultrasonic measurements previously performed on a non-deuterated sample. It can be noticed that, within the experimental error, the IXS and BLS values of v(T) are the same in the glassy phase, i.e. for T < 180K, while for higher temperatures, the BLS sound velocity shows a steeper temperature dispersion related to the structural relaxation. IXS data are less sensitive to this relaxation and can be used as a good estimation for the unrelaxed sound velocity v,.

This is also demonstrated by the comparison with the values of v_∞ previously obtained by fitting the BLS spectra. The IXS data are obviously more reliable since they come from a measure rather than from a fitting procedure, and since they correctly extrapolate towards the low temperature BLS data, where the structural relaxation does not effect the velocity value extracted from the BLS spectra. The data of v_∞ reported here promise to be decisive to obtain the temperature behavior of the relaxation time and relaxation strength from BLS spectra of polybutadiene, quantities which are relevant to the experimental test of the predictions of the mode couplig theory (MCT). This kind of analysis is currently in progress.
Among the predictions of the MCT there is the one on a singular behavior of the non-ergodicity factor at T_c, i.e. of the strength of the structural relaxation. A direct access to this quantity can also be gained from IXS measurements as the ratio between elastic to inelastic intensities. The temperature behavior of this quantity is reported in Fig.3 for three different Qs. Although the data are scattered due to low statistics, some change in the temperature dependence can be seen around the glass transition. In particular, the ratio decreases for increasing temperature at low T and tends to become constant above the transition, which is a behavior consistent with the predicitons of the theory.
Finally, it must be mentioned that the present experiment was part of a joint ESRF-ILL research on the dynamics of polybutadiene. A more comprehensive report on the results obtained by neutron scattering measurements performed at IN5 will be submitted to the ILL. It can be useful here to summarise the most relevant result: Neutron measurements evidenciate a strong Umklapp scattering in PB which extends down to 4 nm^{-1}. This result leeds to infer that this Umklapp signal should also be seen in IXS at higher Q values, perhaps modified by the differences between x-rays and neutrons atomic scattering amplitudes.
In summary, our measurements show the ability of IXS to determine the unrelaxed sound velocity of glass forming polymers. Moreover, IXS spectra have been used to estimate the temperature behavior of the MCT non-ergodicity factor. The results obtained strongly encourage a more thorough study in the vicinity of the critical temperature of this system: this should allow to better reveal the singularity predicted by the MCT. Moreover, measurements at higher Q-values should be done to investigate the existence of Umklapp signal already observed by neutron scattering measurements on this sample.

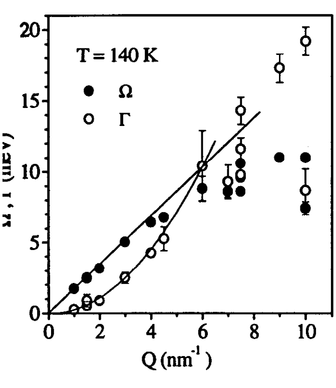

Figure 1

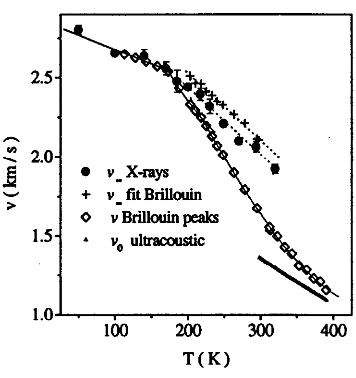

Figure 2

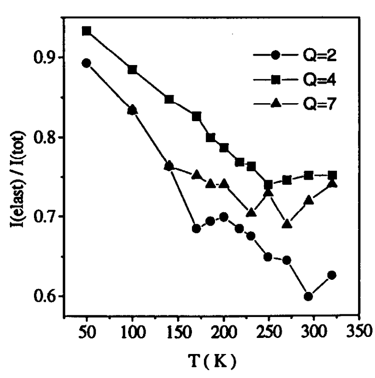

Figure 3