



DETERMINATION OF THE INFINITE FREQUENCY SOUND VELOCITY IN THE GLASS-FORMER *ORTHO*-TERPHENYL

G. MONACO¹, C. MASCIOVECCHIO², G. RUOCCO¹ AND F. SETTE²

¹ UNIV. DI L'AQUILA AND ISTITUTO NAZIONALE DI FISICA DELLA MATERIA, L'AQUILA (ITALY)

² ESRF, EXPERIMENTS DIVISION

The high-frequency dynamics of the glass-former *ortho*-terphenyl is studied here in the glass transition region. Inelastic X-ray Scattering (IXS) is used to determine the dynamic structure factor $S(Q, \omega)$ in presence of relaxation processes, in a Q regime which is not accessible to other techniques, namely Brillouin Light Scattering and Inelastic Neutron Scattering.

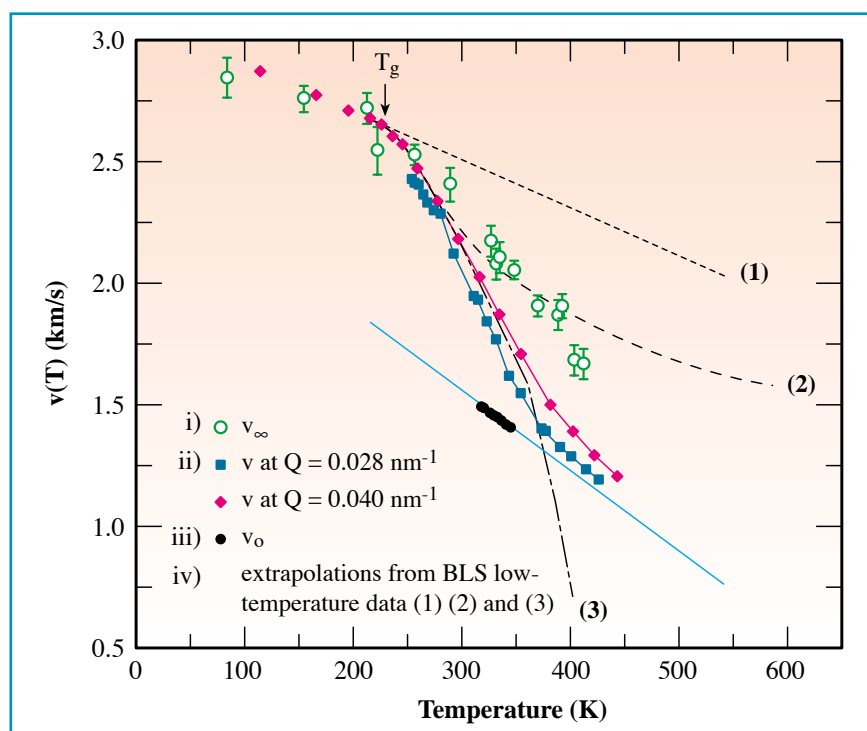
Simple liquids undergoing the glass transition, like the organic glass-former *ortho*-terphenyl studied in the present paper, are characterized by the so-called α (structural) relaxation, whose timescale $\tau_\alpha(T)$ is strongly temperature dependent. Due to this relaxation, the speed of the longitudinal waves propagating at temperature T with wavevector Q , $v(Q, T)$, presents a transition from a low-frequency ($\omega = 2\pi\nu \ll 1/\tau_\alpha(T)$) adiabatic value, $v_0(T)$ to the high frequency ($\omega = 2\pi\nu \gg 1/\tau_\alpha(T)$) purely elastic value, $v_\infty(T)$. The specific distribution of relaxation times, as well as $v_0(T)$ and $v_\infty(T)$, are the essential parameters governing the dispersion of the sound waves excitation energies and, therefore,

their experimental determination has always received great attention. The condition $\omega \ll 1/\tau_\alpha(T)$ can be met by low frequency spectroscopies, like ultrasonic ones; thus $v_0(T)$ can be measured in a wide temperature range, from the liquid phase almost down to the calorimetric glass transition temperature, T_g . The determination of $v_\infty(T)$ is much more difficult. Brillouin Light Scattering (BLS) methods detect the sound wave speed at about 10 GHz, and correspondingly determine $v_\infty(T)$ only in the glass and in the deeply under-cooled liquid where $\tau_\alpha \geq 10^{-11}$ s. The lack of information on $v_\infty(T)$ in the liquid region is currently covered by different extrapolation schemes of the low-temperature data.

The recent development of the Inelastic X-ray Scattering (IXS) technique allows to study the dynamic structure factor $S(Q, \omega)$ in a Q regime accessible neither to BLS (for which $Q \approx 0.05 \text{ nm}^{-1}$) nor to Inelastic Neutron Scattering (due to kinematic limitations). This gives the possibility to measure the dispersion relation of acoustic excitations, $\Omega(Q)$, up to frequencies where the condition $\Omega(Q) \gg 1/\tau_\alpha$ is verified in the liquid state. The quantity $v_\infty(T)$ can then be derived as the $Q \rightarrow 0$ limit of $v(Q, T) = \Omega(Q)/Q$.

In this study we performed Q - and T -dependent IXS measurements aiming to determine the temperature dependence of $v_\infty(T)$ in *ortho*-terphenyl, a typical fragile

Fig. 1: Temperature dependence of different sound velocities in *ortho*-terphenyl:
 i) $v_\infty(T)$ derived from IXS measurements (open circles). ii) $v(T)$ derived from BLS measurements at $Q \approx 0.028$ (full squares) and 0.04 (full diamonds) nm^{-1} .
 iii) $v_0(T)$ derived from ultrasonic measurements (dots) and linearly extrapolated on a wider temperature region (solid line). iv) $v_\infty(T)$ extrapolated in the liquid from low-temperature BLS data assuming: (1) a linear extrapolation from the glass values of v_∞ (dotted line), (2) a linear dependence of the compliance $J_\infty(T)$ (dashed line) extrapolated from its values at temperatures slightly above T_g , and (3) a linear dependence of the elastic modulus $M_\infty(T)$ (dot-dashed line) extrapolated from its values at the same temperatures as $J_\infty(T)$.





glass forming system, in a temperature range covering the glass and the liquid phases. The values obtained for $v_\infty(T)$ are shown as open circles in Figure 1 and are compared to literature data of : i) $v_0(T)$ as measured by ultrasonic methods (dots) and ii) $v(Q, T)$ as measured by BLS at $\approx 0.04 \text{ nm}^{-1}$ (full diamonds) and $\approx 0.028 \text{ nm}^{-1}$ (full squares). The BLS results report the typical S-shape evolution due to the structural α relaxation, and $v(Q, T)$ makes the transition from v_∞ in the glass to a plateau in the high-temperature liquid. In the glass, where $v(Q)$ is expected to coincide with $v_\infty(T)$, there is good agreement between the IXS and BLS $v(Q)$ values. Such a result implies that in the glass region, the procedure used to extract v_∞ from the IXS data is reasonable.

Therefore, there is confidence that also in the liquid, i.e. in a temperature range not accessible before, the values obtained for v_∞ are meaningful. They are reported in Figure 1 together with three extrapolation schemes, typically utilized to infer v_∞ from low-temperature BLS data. The dotted line is obtained from v_∞ in the glass, linearly extrapolated in the liquid. Compared to the IXS determination, v_∞ is overestimated. The dashed line is obtained assuming a linear temperature dependence of the compliance $J_\infty \approx 1/\rho v_\infty^2(T)$, where ρ is the mass density. The dot-dashed line, on the other side, assumes a linear temperature dependence to the elastic modulus $M_\infty \approx \rho v_\infty^2(T)$. These last two extrapolations give both a good estimate of $v_\infty(T)$ at temperatures slightly higher

than the glass transition temperature T_g . Only the estimate based on the temperature dependence of J_∞ , however, is capable to give $v_\infty(T)$ values very similar to those obtained by IXS in the whole considered temperature range.

In summary, we have shown that it is possible to determine the infinite frequency sound velocity $v_\infty(T)$ from the high frequency dispersion relation measured by IXS. The knowledge of this parameter will allow better modelling of the $S(Q, \omega)$ in presence of relaxation processes, and, therefore, to get more reliable values of the relaxation times. ■

REFERENCE

G. Monaco, C. Masciovecchio, G. Ruocco and F. Sette, *Phys. Rev.* 80, 2161 (1998)