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Fast dynamics of $Na_{0.5}Li_{0.5}PO_3$ observed by coherent neutron scattering

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Abstract

Experimental results obtained by coherent neutron scattering with the time of flight spectrometer MIBEMOL on the less-fragile glass-forming liquid $Na_{0.5}Li_{0.5}PO_3$ are presented. A qualitative discussion of the susceptibility spectra within the framework of the simplest version of the mode-coupling theory locates a possible crossover temperature T_c near 620 K ($\sim 1.2T_g$) close to the temperature of the decoupling phenomena of the relaxation time scales observed by ^{31}P NMR.

Keywords: Coherent scattering; Glass transitions; Mode coupling; Time of flight

During the last decade, the glass transition phenomena has known a renewed interest triggered in particular by the achievement of the mode-coupling theory (MCT) [1]. In recent neutron scattering experiments [2–4], some predictions of the MCT have been verified in particular above T_c for a class of glass-forming liquids which are called fragile liquids. However, it is not clear up to now whether this dynamic instability is also relevant in less-fragile systems which are characterized by a smoother temperature dependence of the structural relaxation time scale and a stronger influence of the vibrational excitations on the dynamics.

So in order to approach this problem, we are studying the dynamical behavior of an alkali phosphate glass-forming liquid [5]. $Na_{0.5}Li_{0.5}PO_3$ ($T_g = 515$ K, $T_m = 749$ K) forms long twisted chains of covalent-bonded PO_4 tetrahedra. $Na_{0.5}Li_{0.5}PO_3$ occupies an interesting position in known glass-forming systems between strong and fragile liquids as glycerol or $ZnCl_2$. Furthermore, it is rather easy to prevent the supercooled liquid from crystallizing in the whole temperature range between T_g and T_m thus over more than 200 K. Neutron scattering experiments have been carried out on the time of flight (TOF) spectrometer MIBEMOL at the Laboratoire Léon Brillouin with a closed hollow niobium cell in the temperature range 300–773 K ($\lambda_i = 6.2$ Å, FWHH $= 84$ μeV). Using pure ^7Li for the synthesis, scattering by $Na_{0.5}Li_{0.5}PO_3$ is mainly coherent (93%) and as 87% of the signal is due to the PO_4 tetrahedra, neutron scattering is a very convenient tool to study the dynamics of the phosphate chains. After usual corrections (detector efficiencies, empty can), the TOF spectra were converted in $S(q, \omega, T)$. Because of the high sample transmission (90%), multiple scattering corrections were not applied.

As the structural relaxation (α-process in the framework of the MCT) remains essentially not

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resolved with the chosen experimental setup, the spectra obtained from MIBEMOL show mainly two contributions which have to be regarded as specific for disordered materials. The first one is a broad asymmetric component, the so-called Boson peak with a maximum located around 6.4 meV in this system at room temperature as observed also in other phosphate glasses by Raman scattering together with optical modes at higher energies (40 meV) [6]. The second component observed at lower energies seems to be quasielastic (MCT β-process) and its intensity increases with temperature stronger than the Bose factor, especially above the glass transition temperature T_g.

The fact that the two relaxational processes described by the MCT (α and β relaxations) remain strongly separated in frequency over the whole temperature range of supercooling has enabled us to test some of the basic features of the idealized MCT in the β regime. First, the factorization property of the dynamic susceptibility $\chi''(q, \omega, T) = S(q, \omega, T)/n(\omega, T)$ in q and ω functions $\chi''(q, \omega, T) = \chi''(\omega, T)\, S^{\text{inel}}(q)$ predicted by the MCT has been verified in the whole temperature range investigated. It has been found that the data for different q overlap within experimental accuracy not only in the β regime but also for the Boson peak. Furthermore, the inelastic structure factor $S^{\text{inel}}(q)$ deduced from this factorization is not temperature dependent. Although the verification of this factorization property is a prerequisite condition for any mode coupling analysis in the β regime, it does not exclude other interpretations of the data. In particular, the observed similar q dependence of the dynamical structure factor for the Boson peak and the quasielastic region does not preclude from a vibrational origin for the latter. Using the convolution model [7] which associates these two processes, very good description of the spectra have been achieved. However, the physical meaning of the involved parameters is not obvious.

As shown in Fig. 1, the susceptibility spectra obtained are almost flat with a prominent contribution of the Boson peak at high energy. It results in distorted spectra with respect to the MCT scenario as in the case of other non-fragile systems actually studied. So in order to derive the value of an eventual crossover temperature T_c, the only prediction

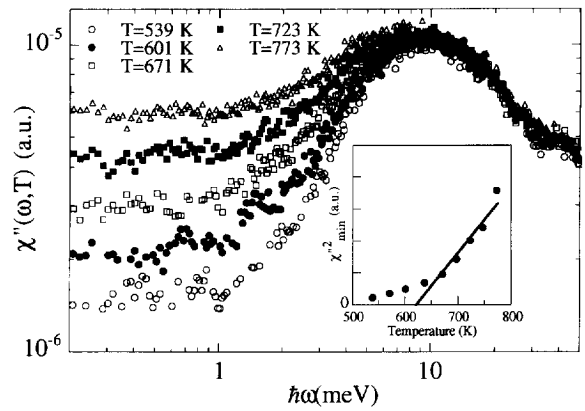

Fig. 1. Susceptibility spectra at some temperatures above T_g obtained by coherent neutron scattering with the TOF spectrometer MIBEMOL on $Na_{0.5}Li_{0.5}PO_3$. At high energy, the Boson peak is clearly seen whereas, at lower energy, a quasielastic component (MCT β process) dominates the spectra. The inset shows the temperature dependence of the susceptibility plateau height deduced from these susceptibilities leading to a possible crossover temperature $T_c \sim 620$ K in good agreement with the decoupling phenomena of the relaxation timescales observed by ^{31}P NMR [8].

of the MCT which can be easily tested is the temperature dependence of the susceptibility plateau height $\chi''^2_{\text{min}} \propto T - T_c$ as shown in the inset of the figure. It leads to a possible crossover temperature T_c near 620 K ($\sim 1.2 T_g$) close to the temperature of the decoupling phenomena of the relaxation timescales observed by ^{31}P NMR [8] as in the case of fragile liquids.

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