

High frequency collective dynamics in liquid potassium

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Abstract

We investigated by inelastic X-ray scattering the dynamical properties of molten potassium in a wide range of momentum transfer, Q , from 1 nm^{-1} up to the main peak of the structure factor $Q \approx 17 \text{ nm}^{-1}$. The observed increase of sound velocity in the low Q region ($1 < Q < 3 \text{ nm}^{-1}$), has been described within a model characterized by two distinct relaxation processes for the collective dynamics. The obtained results are discussed and compared with those from previous neutron scattering experiments. In particular, we associate the speed-up of the sound velocity to the 'instantaneous' disorder of the liquid as opposed to the argument, supported by some neutron scattering studies, of a transition from a liquid to solid like response of the system.

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1. Introduction

The study of the microscopic dynamics of simple liquids received in the last few years has renewed interest, particularly from the point of view of experimental investigations [1–10]. In fact, thanks to the parallel application of similar spectroscopic techniques, namely inelastic X-ray scattering (IXS) and inelastic neutron scattering (INS), it is possible today to go deeper into the investigation of the microscopic mechanisms controlling the dynamical behavior of a simple liquid, being well supported by experimental results.

In particular, one of the preferred classes of systems, selected to study collective and single particle dynamics, is represented by the alkali metals. These liquids, indeed, present well-pronounced inelastic features in their density fluctuation frequency spectra, even at length-scales far

away from the hydrodynamic limit and comparable with the average distance between the particles.

Looking at collective properties, IXS spectroscopy technique has been massively adopted in the recent past [1,7–9] in order to get an insight into the ultimate mechanisms ruling the high frequency dynamics. In the present study we report an investigation of the dynamics of liquid potassium in a wide wave vector–energy (Q – E) region. We discuss possible models to describe the collective dynamics through a comparison with similar INS data recently published [3,4].

More specifically, several recent experimental investigations of liquid dynamics have put forward the crucial role played by the memory function formalism [1,5,6,9,10], a framework tailored to account for the relaxation spectra of the density fluctuations over an extended wave-length energy range. Most of the work of the present study on liquid potassium has been devoted to understand how the evolution of the $S(Q, \omega)$, varying Q values, could be described with a suitable memory function model.

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On a theoretical ground, the hydrodynamic equation of a simple fluid predicts the presence of two distinct relaxation processes for the decay of the density fluctuations [10,11], one associated with the coupling of density and temperature fluctuations and another stemming from the viscous relaxation.

Starting from these indications on the nature of the relaxation processes in the hydrodynamic limit, we test the soundness of a model where the viscous relaxation proceed through two distinct channels [13–16] which are active on quite different time scales. This model, already applied in other systems [1,5,6,9,10], depicts a microscopic scenario where the atoms experience, in the sub-pico-seconds time region (τ_μ), the interaction with the surrounding neighbors. This relaxation process has been termed as microscopic and instantaneous, in contrast to the structural relaxation time that acts on a longer timescale and controls the rearrangement of the instantaneous atomic equilibrium configuration.

The purpose of the present paper has been first to observe the evolution of the dynamics of the liquid at different lengths, studying the dominant excitation frequency of the $S(Q, \omega)$. Then we proceed in the description of the microscopic mechanism of the liquid exploiting the physical meaning of the parameters involved in the model. Our attention has been focused on the low Q region where INS experiments are affected by well-known kinematics limitations.

Previous INS investigations of liquid potassium have been analyzed with both an empirical [3] and memory function framework [4], similar to the one adopted in previous IXS studies [1,5,6,9,10]. The values of speed of sound deduced from the INS experiments exceed the hydrodynamic value, and such phenomenon has been interpreted as reminiscence of the crystal like sound propagation. This explanation, which stems from an interpretation of the relaxation dynamics in terms of a liquid to solid like transition, is here discussed also in view of several other results reported in other liquids and (numerically) supercooled and glassy metals (Li, Na, Al, Ga) [1,5,6,9,10,17,18].

2. Experiment

We report here the determination, by means of the IXS spectroscopy, of the dynamic structure factor of liquid potassium in the Q range from 1 nm^{-1} to 16 nm^{-1} . The experiment has been performed at the high-resolution beam line ID16 at the European Synchrotron Radiation Facility, in a fixed exchanged wave-vector configuration. Thanks to the presence of a five analyzers bench, we collected simultaneously five different values of Q , then we have been able to map the planned Q range with a satisfying accuracy spending a reasonable time for the data acquisition. For all spectra, we performed an energy scan (–50 to 50 meV) that took about 300 min and was repeated to collect 300 s per point of total integration time. During the measurements we fixed the temperature of the potas-

sium slightly above the melting point at $T = 343 \pm 1 \text{ K}$, keeping the sample under vacuum. The sample holder for liquid potassium was made out of two pieces of austenitic steel which was able to conform the liquid to be confined with a thickness of 1 mm. Such length corresponds to the absorption length of the potassium at the energy of the incident beam. The energy resolution used during the measurements, 1.5 meV (FWHM), corresponds to the chosen reflection of the silicon analyzer (11 11 11) for an incident energy of the beam of 21 748 eV. In Fig. 1, we show the collected raw spectra in all the explored Q range. It is evident, even at this level, the presence of a dispersion behavior of the inelastic structure of the spectra; the dispersion reaches the maximum at $Q = 10 \text{ nm}^{-1}$ and progressively decreases when Q approaches the main peak of the static structure factor 16 nm^{-1} .

3. Analysis and discussion

In order to quantify the role of the different microscopic mechanisms that drive the dynamics of liquid potassium, we modelled the dynamical $S(Q, \omega)$ using the memory function formalism. Starting from the generalized Langevin equation, it is possible to write $S(Q, \omega)$ in terms of real ($M'(Q, t)$) and imaginary ($M''(Q, t)$) part of the Fourier–Laplace transform function $M(Q, t)$ [11,12], so that

$$S(Q, \omega) = \frac{S(Q)\pi^{-1}\omega_0^2(Q)M'(Q, \omega)}{[\omega^2 - \omega_0^2(Q) + \omega M''(Q, \omega)]^2 + [\omega M'(Q, \omega)]^2}.$$

Here we have introduced the frequency $\omega_0(Q) = KTQ^2/mS(Q)$ calculated from the liquid static structure factor $S(Q)$, this quantity is directly connected to the generalized isothermal speed of sound through the relationship: $c_t(Q) = \omega_0(Q)/Q$.

The ultimate goal is, of course, to find the most appropriate shape for the $M(Q, t)$, each relaxation process entering the spectrum of the density fluctuation can be taken into account as an additive contribution to the second order memory function.

Starting from the generalization of the hydrodynamic we chose to use a model for the $S(Q, \omega)$ characterized by three distinct channels for the relaxation processes. The first channel arises from the coupling between the density and the temperature fluctuations, while the other two are associated with the viscous relaxation process.

$$M(Q, \omega) = (\gamma - 1)\omega_0^2(Q)e^{-D_T Q^2 t} + \Delta_z^2(Q)e^{-t/\tau_z(Q)} + \Delta_\mu^2(Q)e^{-t/\tau_\mu(Q)}.$$

The specific heat ratio γ and the thermal diffusivity D_T have been deduced from macroscopic data [19].

We use for the thermal contribution to the $M(Q, t)$ the generalized hydrodynamic result, i.e. the usual Debye expression, known as viscoelastic model [20], neglecting any Q dependence of the diffusion coefficient. Moreover, similarly to previous studies on alkali metals [1,5,6,9,10,18], we adopt a two exponential functions

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