The mixed longitudinal–transverse nature of collective modes in water

A Cimatoribus¹,⁴, S Saccani¹,⁴, F Bencivenga¹, A Gessini¹, M G Izzo¹,² and C Masciovecchio¹,³
¹ Sincrotrone Trieste, 34149 Basovizza, Trieste, Italy
² Dipartimento di Fisica, Università degli Studi di Trieste, 34127 Trieste, Italy
³ CRS SOFT-INFM-CNR, Università di Roma La Sapienza, 00185 Roma, Italy
E-mail: andrea.cimatoribus@gmail.com and sebastiano.saccani@gmail.com

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Abstract. We report high-resolution, high-statistics inelastic x-ray scattering measurements of the dynamic structure factor of water as a function of momentum and energy transfer in various thermodynamic conditions, including high-pressure liquid near the melting point, supercooled liquid and polycrystalline ice. For momentum transfer values below 8 nm⁻¹, two collective excitations associated with longitudinal and transverse acoustic modes were observed. Above 8 nm⁻¹, another excitation was detected in the liquid. Comparison with polycrystalline data and molecular dynamics simulations suggests that this mode is related to longitudinal–transverse mixing of mode symmetry.

Contents

1. Introduction 2
2. Experimental setup and fitting procedure 3
3. Experimental results 5
4. Conclusions 8
Acknowledgments 9
References 9

4 Authors to whom any correspondence should be addressed.
1. Introduction

Water is a system that has been widely studied because of its peculiar thermodynamic anomalies whose origin is not yet fully understood and because of its prominent role in several fields of applied and life sciences [1, 2]. One of the challenges that scientists are facing concerns the description of the high-frequency collective dynamics [3–17]. Notwithstanding the considerable amount of effort, a commonly accepted scenario is still missing.

The existence of a single longitudinal acoustic (LA) mode at low momentum transfer ($Q \lesssim 0.03 \text{ nm}^{-1}$) has been demonstrated, propagating at a velocity equal to the adiabatic one ($c_S$) [18]. At higher $Q$ values, a large increase of the speed of sound with respect to $c_S$ (positive sound dispersion) is accompanied by the emergence of a weakly dispersive feature, at energies below the LA mode [6]. Whereas positive sound dispersion is fully accounted for by the viscoelastic theory of liquids [15, 19, 20], the nature of the second excitation is much more debated. At first, it was simply identified as the LA mode [3]. Molecular dynamics (MD) simulations showing the existence of this excitation date back to 1974 [21]. Later on, transverse optic nature was inferred based on inelastic x-ray scattering (IXS) experiments [6, 22] and further MD simulations [23–25]. Successive experimental and theoretical investigations pointed out the transverse acoustic (TA) character of this excitation [16, 26, 27]. Afterwards, the prevalent acoustic character of this mode was challenged again by neutron scattering experiments [10, 17].

Therefore, the first purpose of the present work is to identify the nature of the second excitation. To achieve this goal, we analyzed the lineshape of IXS spectra collected with an unprecedentedly high statistical accuracy in the region of exchanged momentum ($Q$) and energy ($\hbar \omega$) where the aforementioned phenomenology shows up. The sample was probed in different thermodynamic conditions, including the supercooled liquid phase, where experimental data on the high-frequency collective dynamics are lacking. IXS spectra of liquid and supercooled water were analyzed within the viscoelastic approach [19]. According to this framework, the dynamic response of the liquid is similar to that of the solid as long as the inverse structural relaxation time is much shorter than the frequencies of probed excitations. In the case of water, this condition is essentially met in the investigated thermodynamic range, particularly close to or below the melting point [6, 8, 9, 13]. Consequently, the analysis of IXS spectra from supercooled and polycrystalline water (the latter data were collected during the same experiment) in the same thermodynamic conditions allowed us to establish a closer comparison between the liquid and solid phases.

Our data were also compared to the literature results and allowed us to unambiguously assess the TA nature of this low-frequency mode (LFM) for $Q \lesssim Q_{\text{MAX}}/2$, where $Q_{\text{MAX}} \approx 8–9 \text{ nm}^{-1}$ is the $Q$ value corresponding to the first sharp diffraction peak in the static structure factor ($S(Q)$). Above $Q_{\text{MAX}}$, the prevalent TA nature of LFM cannot be decided anymore. Furthermore, a new high-frequency mode (HFM) is found in the liquid. We interpret this feature as likely due to intramolecular motions of mixed longitudinal–transverse symmetry. Even if we cannot exclude that the HFM has already been recorded in previous IXS experiments, its existence has never been reported. This is most likely due to the fact that the HFM is hardly discernible from the background in view of its very low cross section, and former investigators did not have the capability to analyze data of such high statistics, nor to study the supercooled liquid phase where, indeed, the HFM is slightly more evident.
Table 1. Thermodynamic point investigated in the present work. The probed $Q$-range and the employed sample environment were reported as well.

| $T$ (K) | $P$ (Bar) | $Q$ (nm$^{-1}$) | Phase   | Setup   |
|---------|-----------|-----------------|---------|---------|
| 263     | 1.2       | 2–18            | Supercooled | Drop    |
| 263     | 1         | 2–14            | Polycrystalline | Drop    |
| 263     | 3000      | 2–14            | Liquid  | HP      |
| 278     | 1         | 2–14            | Liquid  | HP      |
| 278     | 3000      | 2–14            | Liquid  | HP      |
| 288     | 3000      | 2–14            | Liquid  | HP      |
| 353     | 3000      | 2–14            | Liquid  | HP      |

2. Experimental setup and fitting procedure

Measurements were carried out at the IXS beamline (ID28) at the European Synchrotron Radiation Facility in Grenoble. ID28 was operated at the Si(11,11,11) configuration, delivering a total energy resolution of about 1.5 meV (full-width at half-maximum). Further details of the instrument can be found elsewhere [28]. The dynamical structure factor, $S(Q, \omega)$, was measured in the $Q$ range of 2–18 nm$^{-1}$ and in the $\bar{\hbar}\omega$ range of ±40 meV. The typical acquisition time for a set of IXS spectra was about 12–16 h. Table 1 reports the thermodynamic and kinematic points investigated in our experiment.

Two different sample environments were employed. The first one, suitable for undercooling, has been derived from a setup used for studying homogeneous nucleation [29] (Drop). In this case, the sample was a droplet of high-purity water of approximately 3 mm diameter. It was kept on a 100 $\mu$m-thick hydrophobic glass, sealed in a 5 × 2 cm$^2$ (diameter × height) chamber in a helium atmosphere at 1.2 bar, in order to prevent contamination from the atmosphere. This newly developed device prevented crystallization in the supercooled phase at 263 K for several hours, thus enabling IXS measurements. This apparatus was also used to collect data on polycrystalline ice $I_h$ at the same temperature. The second setup is a high-pressure (HP) apparatus already used to carry out several IXS experiments [8, 16, 30].

Data were analyzed using the viscoelastic approach [19], which has already been successfully employed in the interpretation of high-frequency dynamics of liquid water and other liquids [8, 9, 13, 16, 19, 30, 31]. The expression for $S(Q, \omega)$ can be obtained within the memory function formalism. Memory function theory is used to derive a generalized Langevin equation for the density correlation function, $F(Q, t)$:

$$\frac{\partial^2 F(Q, t)}{\partial t^2} + \omega_0^2 F(Q, t) + \int_0^t m(Q, t - t') \frac{\partial F(Q, t')}{\partial t'} dt' = 0,$$

where $\omega_0$ is the frequency of sound excitation in the hydrodynamic ($Q \to 0$) limit and $m(t)$ is the memory function. The Fourier transform of $F(Q, t)$, i.e. $S(Q, \omega)$, then reads:

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

where $M'(Q, \omega)$ and $M''(Q, \omega)$ are the first and second derivatives of the memory function, respectively.
where \( M'(Q, \omega) \) and \( M''(Q, \omega) \) are the real and imaginary parts of the Fourier transform of \( m(Q, t) \). In the case of water, \( m(Q, t) \) can be approximated as [8, 13, 16]:

\[
m(Q, t) = (\gamma(Q) - 1)(c_1(Q) Q)^2 e^{-D_T(Q)Q^2} + \Delta_\omega(Q)^2 e^{-1/\tau_\omega(Q)} + 2 \Gamma_\mu(Q) \delta(t).
\]  

(3)

In the first term on the right-hand side of (3), \( c_1(Q) \), \( \gamma(Q) \) and \( D_T(Q) \) are the \( Q \)-dependent generalizations of the isothermal speed of sound, specific heats ratio and thermal conductivity, respectively. In the second term on the right-hand side of (3), \( \Delta_\omega(Q) \) and \( \tau_\omega(Q) \) are the structural relaxation strength and time, while in the third term, \( \Gamma_\mu(Q) \) represents the strength of the instantaneous relaxation [8, 9, 13, 16, 19]. It is also useful to introduce the LA frequency in the elastic regime: \( \omega_\infty = \sqrt{\omega_0^2 + \Delta_\omega^2} \). In order to account for the aforementioned weakly dispersive LFM and HFM, we added two damped harmonic oscillator (DHO) functions, following the idea of [16]:

\[
S(Q, \omega) = \frac{I_L}{\pi} \frac{\omega_0^2 M'(Q, \omega)}{[\omega_0^2 - \omega^2 - \omega M''(Q, \omega)]^2 + [\omega M'(Q, \omega)]^2} + I_T \frac{\Omega_T^2 \Gamma_T}{(\Omega_T^2 - \omega^2)^2 + (\omega \Gamma_T)^2} + I_{HFM} \frac{\Omega_{HFM}^2 \Gamma_{HFM}}{(\Omega_{HFM}^2 - \omega^2)^2 + (\omega \Gamma_{HFM})^2},
\]

(4)

where \( \Omega_T \) and \( \Gamma_T \) are the characteristic frequency and damping of LFM, while \( I_T \) and \( I_L \) are scaling factors. For \( Q \approx 8 \text{ nm}^{-1} \), we were able to detect the HFM, empirically accounted for in the model function by adding a further DHO function, denoted in (4) by the suffix ‘HFM’. The resulting \( S(Q, \omega) \) has to be multiplied by the weighting factor:

\[
w(\omega, T) = \frac{\hbar \omega}{k_b T}[1 - e^{-\hbar \omega/k_b T}]^{-1}
\]

(5)

in order to account for occupation of the different vibrational levels. Finally, a convolution with the experimental resolution function \((R(\omega))\) and the addition of a flat background \((B)\) are needed to obtain the complete fitting function:

\[
I(Q, \omega) = R(\omega) \otimes [w(\omega, T) \times S(Q, \omega)] + B.
\]

(6)

Fits are based on \( \chi^2 \) minimization; the description of the experimental data was always good, with reliability better than 0.99. It must be stressed that all lineshape parameters were always left free to vary without constraints. The results using the model currently described are consistent with those from a model with three DHOs.

In figure 1, selected IXS spectra of polycrystalline and liquid water are shown on a logarithmic scale, together with the corresponding best fit results. In the liquid, besides the dominant LA mode, we observe that the low-energy excitation is properly fitted (figure 1, middle and bottom panels). Above \( 8 \text{ nm}^{-1} \), the new HFM is visible at \( \approx 30 \text{ meV} \). This excitation has never been experimentally observed before and is present both in the supercooled phase and in the HP liquid above the melting point. In the spectra from the polycrystal, in the first pseudo-Brillouin zone (BZ) (top right) a single LFM is observed below the dominant LA branch. In the second pseudo-BZ (top center and left), a double LFM feature is evident. Moreover, at larger energy transfers, the relic of the LA branch is accompanied by the rise of a new HFM in the range 30–35 meV.

To support the consistency of the analysis based on (4), data above \( Q \approx 8 \text{ nm}^{-1} \) were also fitted without the HFM term. As far as the viscoelastic parameters are concerned, both approaches give very similar results. The major difference found was in the parameter \( \omega_0 \), which
Figure 1. Representative IXS spectra from polycrystalline ice at $T = 263$ K (top panels), from supercooled liquid water at $T = 263$ K (‘drop’ setup, medium panels) and from liquid water at $P = 3$ kbar and $T = 288$ K (‘HP’ setup, bottom panels), at $Q = 6.5$ nm$^{-1}$ (left panels), 11 nm$^{-1}$ (central panels) and 14 nm$^{-1}$ (right panels). Empty circles are experimental data; the thick solid line is the best fit results. The dash–dot–dot line is the viscoelastic function accounting for LA and quasi-elastic contributions. The dash–dot line marks low-energy modes and the dashed line is a newly observed high-energy excitation.

is somewhat smaller. To investigate this difference, we compared the obtained results with the expected trend, i.e.

$$\omega_0(Q, T, P) = \frac{c_s(T, P)Q}{\sqrt{S(Q)/S(Q = 0)}}, \quad (7)$$

where $c_s$ is the adiabatic speed of sound, calculated from the well-known equation of state of water, whereas $S(Q)$ was measured contextually to the same experiment directly using the ID28 spectrometer. We observe that (4) with the HFM term gave slightly more consistent results, as can be appreciated by inspecting figure 2, which reports the case of water at 278 K and 3 kbar.

3. Experimental results

The left panels of figure 3 report the $Q$-dispersion relation of characteristic frequencies of the various modes, as extracted from IXS spectra using (4). In order to compensate for differences...
in density, the $Q$-axis was normalized by the measured value of $Q_{\text{MAX}}^5$. In the panel referring to the supercooled liquid, the solid line is the $Q = 0$ extrapolation of sound velocity obtained from a sine fit of LFM for $Q < 12 \text{ nm}^{-1}$, which gives $c_{\text{LFM}} = 1800 \pm 100 \text{ m s}^{-1}$. This estimate of the sound velocity for the LFM in the supercooled case is just below the transverse sound velocity of the polycrystal, calculated from both the Voigt (1947 m s$^{-1}$) and Reuss (1965 m s$^{-1}$) averages [33, 34]. In the calculation, elastic constants measured by Stephens [35] were used. Furthermore, the LFM in the liquid follows the expected dispersion for an acoustic mode, that is, $\omega(Q) \propto Q \times S(Q)^{-1/2}$ [19]. In the right panels of figure 3, the vibrational density of states (DoS) from neutron scattering measurements [11] is displayed. The analysis made by the authors revealed that the energy regions characteristic of LFM (in 8–14 meV) in the DoS of the polycrystal can be unambiguously assigned as arising from acoustic modes [11]. Therefore, the comparison between neutron scattering results and the water and polycrystalline data strongly supports the TA nature of LFM up to $Q/Q_{\text{MAX}} \approx 0.4$.

Moreover, the almost flat dispersion and large intensity of LFM are consistent with the main peak in the DoS, while the intensity decrease of the LA mode at large $E$ is consistent with the plateau between 13 and 27 meV. Similarly, the feature at 25 meV reflects the dispersiveless behavior of the LA branch at high $Q$.

Finally, a comparison of supercooled water data, which are the most sampled dataset, with previous MD simulations is shown in figure 4. The characteristic energies of longitudinal (+) and transverse ($\times$) current correlation functions [27] are displayed. The analogy suggests that the observed LFM is a projection of the transverse current correlation function onto the longitudinal one (see figure 4, bottom panel). In light of these pieces of evidence, we can reasonably conclude that the LFM has a TA nature also in the liquid below 8 nm$^{-1}$. In addition, at higher $Q$ values, MD results highlight the rise of an LFM in the longitudinal current spectrum too.

5 In the case of the polycrystalline sample, $Q_{\text{MAX}}$ is from [32].

New Journal of Physics 12 (2010) 053008 (http://www.njp.org/)
Figure 3. Dispersion of the modes as a function of $Q/Q_{\text{MAX}}$. Full symbols refer to the polycrystal, whereas empty symbols refer to the liquid. On the right part of every panel, DoS on the polycrystal ($Z(E)$) at 270 K and on the liquid at 280 K are displayed, as measured by Dawidowski et al [11]. The branches structure is analogous in the polycrystal and the liquid and is not affected by the sample environment. In the panel referring to the supercooled liquid, the solid line is the extrapolation of TA sound velocity in the $Q=0$ limit. Dashed lines are proportional to $Q \times S(Q)^{-1/2}$ (see text).

However, being almost degenerate with TA modes, the two branches cannot be experimentally discriminated.

Above $Q/Q_{\text{MAX}} \approx 0.4$, the HFM appears in the spectra from the liquid. The rather large dispersion of experimental data together with the impossibility to determine the HFM frequencies at low $Q$ do not allow the unambiguous identification of the $Q$-dispersion relation of such a mode. Therefore, an acoustic or optic-like nature cannot be discriminated on such grounds. However, closer comparison of our data with the DoS reveals that similar vibrational modes are present also in the polycrystal in this energy range. It has been noted that the features of DoS in the polycrystal between 20 and 40 meV are generally discussed in terms of molecular optic modes [11]. In view of the strong analogies mentioned above, it is straightforward to assume that also a mode involving intramolecular motion has a strong resemblance in the liquid and the polycrystal. This holds in particular considering the lack of long-range order of protons in ice $I_h$. Consequently, the HFMs in the liquid should probably be regarded as being of mostly molecular optic nature as well, even though the limited $Q$ range explored in the present work confines the conclusions to qualitative arguments. Looking at the top panel of figure 4, it can
be noted that the experimentally detected HFM rises at the same $Q/Q_{\text{MAX}}$ value where an excitation of similar energy appears in the simulated transverse current correlation function. Thus, it is straightforward to hypothesize that the measured HFM can be ascribed to a projection of the transverse current correlation on the $S(Q, E)$. Moreover, energy separation between HFM and LA branches from the experiment is very close to MD results. The existence of multiple peaks at high $Q$ values is also supported by other simulations [26], carried out using a different intermolecular potential.

4. Conclusions

We reported on and discussed new high-resolution IXS data on water in various thermodynamic conditions, including the supercooled liquid and polycrystalline phases. We provided some evidence of the TA nature of the mode observed in liquid water at frequencies below the dominant LA branch. Above $\approx 8\text{ nm}^{-1}$, we observed a new high-frequency excitation in the liquid phase. It is likely related to intramolecular dynamics and reflects crystalline-like dynamics of the prevalent optic nature. Comparison with MD simulations allows us to assess whether its origin is connected with the transverse current correlation function. The appearance in the density fluctuation spectrum of features from the transverse current correlation function

\[ \text{New Journal of Physics 12 (2010) 053008 (http://www.njp.org/)} \]
fingerprints the mixed longitudinal–transverse nature of microscopic dynamics in water at short wavelengths, and its general resemblance to crystalline dynamics. Since liquids usually show a single branch associated with LA modes, the appearance of multiple excitation branches is likely associated with transverse and longitudinal correlations. This may be a novel manifestation of the unique water structure. Further experimental efforts, aimed at probing dynamics in systems with a water-like local intermolecular structure, are in progress and may shed new light on this topic.

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